

Before the Patent Trial and Appeal Board

IPR2024-00611

U.S. Patent No. 10,471,380

Air Products and Chemicals, Inc.

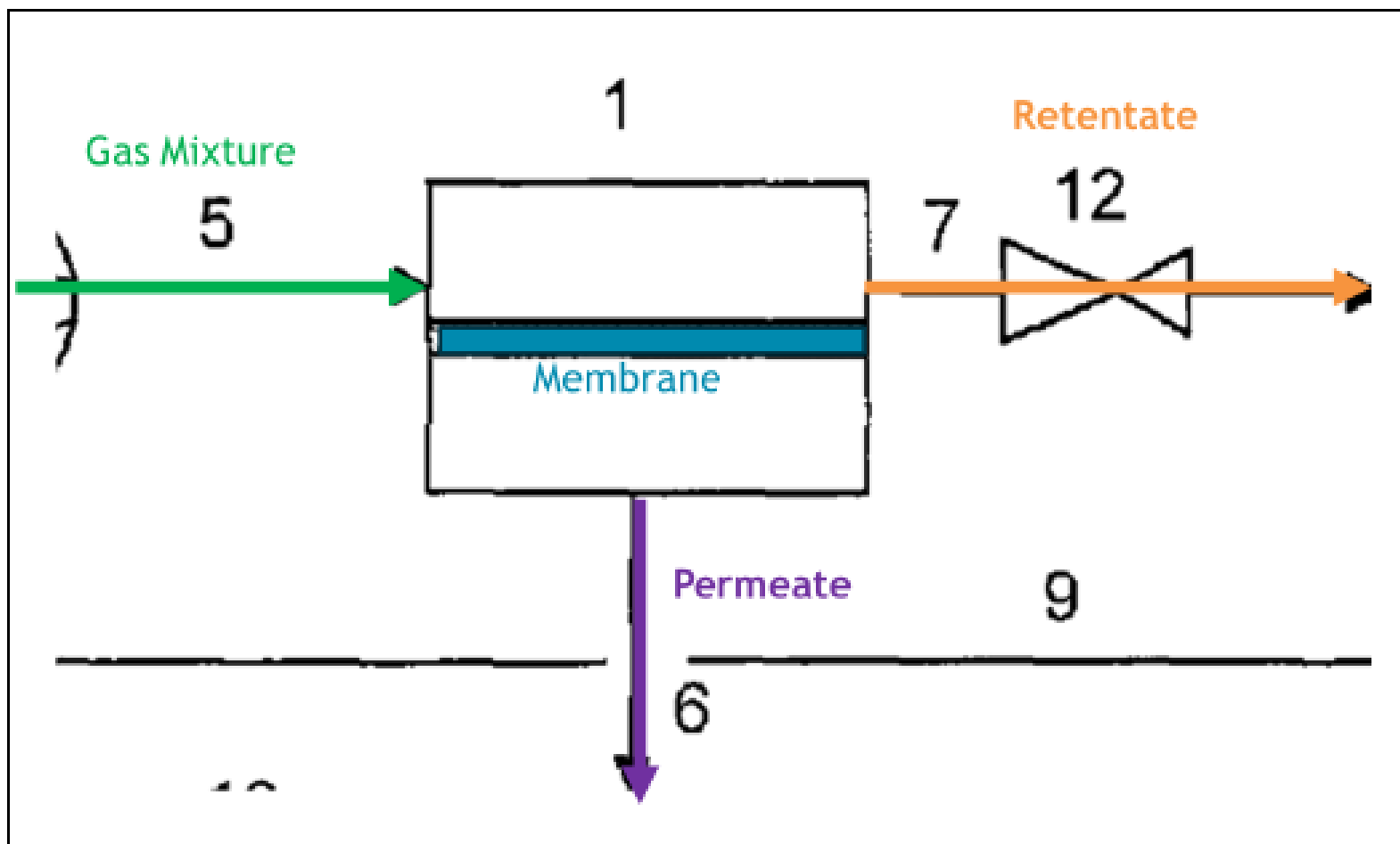
v.

Evonik Operations GmbH and Evonik Corporation

Patent Owners' Oral Argument – May 23, 2025

Overview of the Technology

Background | Gas Membrane Separation



Background | Performance Characteristics

Selectivity. A membrane's *selectivity* is a measure of its preference for permeating one gas relative to another gas. A high selectivity membrane may be more effective at separating one gas from another, but high selectivity generally comes with decreased permeability, increased energy consumption, and higher membrane costs.

Permeance. *Permeance* is a measure of how effectively a certain gas can move through a membrane under a given set of conditions (e.g., temperature and pressure).

Membrane Composition: The *chemical composition*, physical structure, and thickness of a membrane can affect its permeance, selectivity and cost. For example, a membrane made of dense polymers typically offers high selectivity but lower permeability, while a relatively porous membrane may provide higher permeance at the expense of selectivity.

Pressure Ratio. The *pressure ratio* for a membrane separation indicates the difference in pressure between the feed side (where gas is introduced) higher pressure than the permeate side (where gas exits the membrane), and this difference in pressure causes gas to permeate from the feed side to the permeate side. As the pressure ratio increases, more gas overall permeates through (and is separated by) the membrane.

Recycle Volume. The *recycle volume* refers to the amount of gas reintroduced into the feed stream for further separation after the gas has gone through an initial separation process. Recycling of gas streams generally achieves a higher overall purity of a separated gas at the expense of increased energy costs.

Overview of the Priske-'380 Patent



(12) **United States Patent**
Priske

(10) Patent No.: US 10,471,380 B2
(45) Date of Patent: Nov. 12, 2019

(54) **PROCESS FOR SEPARATION OF GASES WITH REDUCED MAINTENANCE COSTS** 53/227 (2013.01); B01D 2053/224 (2013.01); B01D 2317/022 (2013.01); B01D 2317/025 (2013.01)

(71) Applicant: **Evonik Fibres GmbH**, Schoerfling am Attersee (AT)

(72) Inventor: **Markus Priske**, Salzburg (AT)

(73) Assignee: **Evonik Fibres GmbH**, Schoerfling am Attersee (AU)

(*) Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 194 days.

(21) Appl. No.: 15/540,709

(22) PCT Filed: Dec. 22, 2015

(86) PCT No.: **PCT/EP2015/080925**

§ 371 (c)(1),
(2) Date: **Jun. 29, 2017**

(87) PCT Pub. No.: **WO2016/107786**

PCT Pub. Date: Jul. 7, 2016

(65) **Prior Publication Data**

US 2017/0368497 A1 Dec. 28, 2017

(30) Foreign Application Priority Data

Dec. 29, 2014 (EP) 14200384

(51) Int. Cl.
B01D 53/22 (2006.01)
B01D 69/08 (2006.01)
B01D 71/64 (2006.01)

(52) U.S. CL
CPC B01D 53/226 (2013.01); B01D 53/225
(2013.01); B01D 53/228 (2013.01); B01D
69/08 (2013.01); B01D 71/64 (2013.01); B01D

(58) **Field of Classification Search**
CPC B01D 53/22; B01D 53/225; B01D 53/226;
B01D 53/227; B01D 53/228; B01D
69/08; B01D 71/64; B01D 2317/022;
B01D 2317/025
See application file for complete search history.

(56) **References Cited**

U.S. PATENT DOCUMENTS

5,709,732 A *	1/1998	Prasad	B01D 53/226 95/45
6,168,649 B1	1/2001	Jensvold et al.	
6,565,626 B1 *	5/2003	Baker	B01D 53/226 95/47

(Continued)

FOREIGN PATENT DOCUMENTS

(Continued)

OTHER PUBLICATIONS

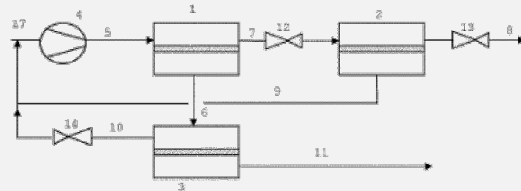
International Search Report and Written Opinion of the International Searching Authority dated Apr. 26, 2016 in PCT/EP2015/080925 filed Dec. 22, 2015.

Primary Examiner — Jason M. Greene
(74) *Attorney, Agent, or Firm* — Oblon, McClelland,
Maier & Neustadt, L.L.P.

(57) ABSTRACT

The invention relates to a specific process and apparatus for separation of gas mixtures with reduced maintenance costs.

23 Claims, 1 Drawing Sheet



Illustrative connection arrangement of several membrane modules according to invention

Patent No.: U.S. 10,471,380

Inventor: Marcus Priske

Assignee: Evonik Operations GmbH

Priority Date: December 29, 2014

Claim 1

1. A method for separating gases from a crude gas stream comprising at least two components, wherein in an apparatus comprising a feed stream separation stage, a retentate separation stage and a permeate separation stage, each stage being a membrane separation stage with gas separation membranes,

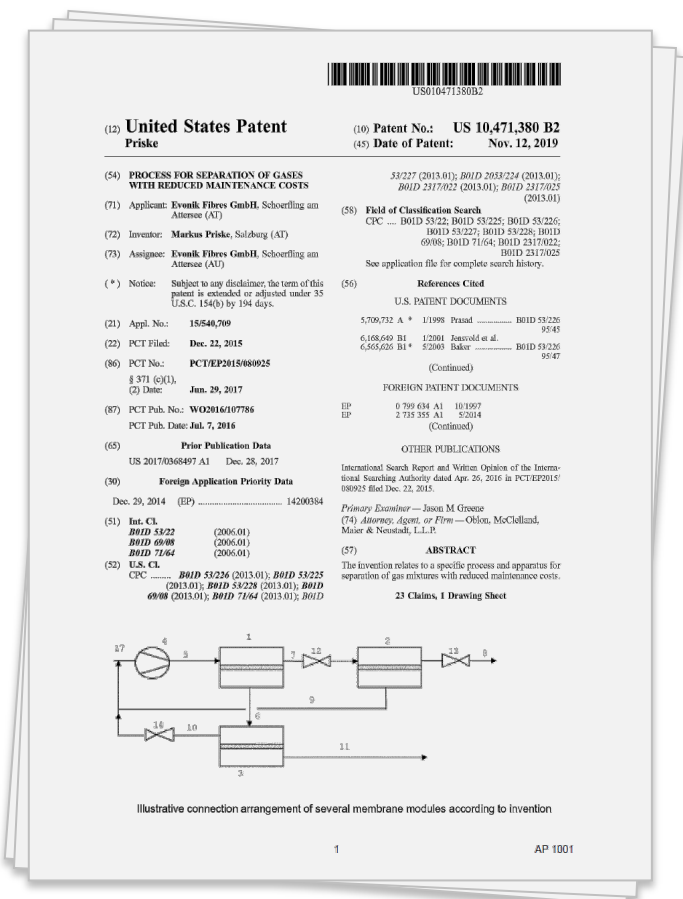
- a) a feed stream is separated in the feed stream separation stage into a first permeate stream and a first retentate stream,
- b) the first retentate stream is separated in the retentate separation stage into a second permeate stream and a second retentate stream,
- c) the first permeate stream is separated in the permeate separation stage into a third retentate stream and a third permeate stream,
- d) the second retentate stream is removed as a first product,
- e) the third permeate stream is removed as a second product, and

f) the second permeate stream and the third retentate stream are combined with the crude gas stream to give the feed stream,

and wherein

- i) the combined gas volume of the second permeate stream and the third retentate stream is from 60 to 100% of the volume of the crude gas stream,
- ii) the total capacity of the gas separation membranes in the retentate separation stage is higher than the total capacity of the gas separation membranes in the feed stream separation stage, the total capacity being measured for nitrogen Grade 4.8 under standard conditions as defined in the description, and
- iii) the quotient of the pressure ratio over the permeate separation stage to the pressure ratio over the feed stream separation stage is in a range of from 0.5 to 8.

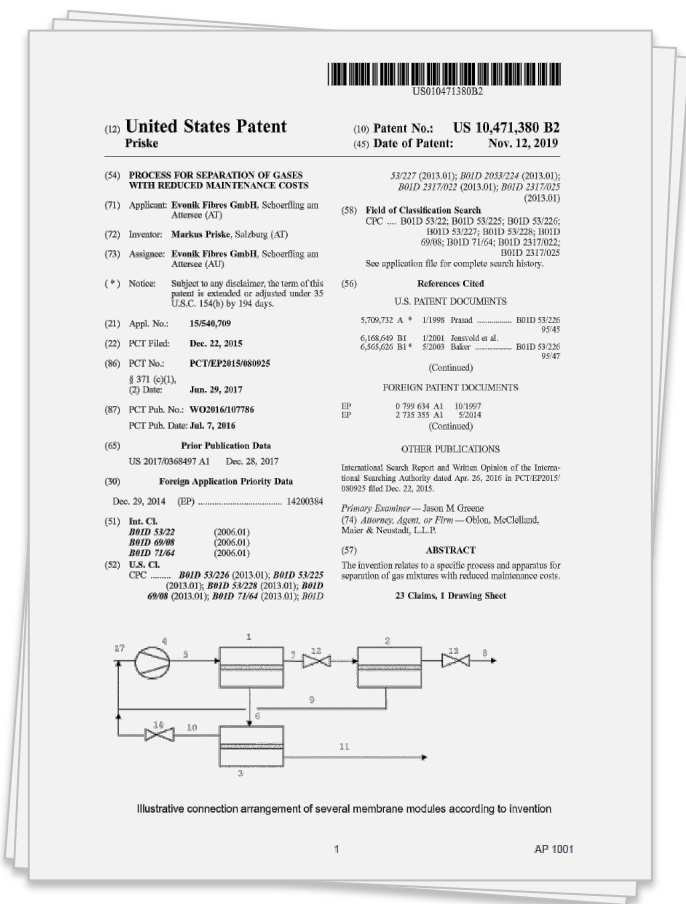
Priske-'380 | Prior Art Focuses on Energy Optimization



It is generally known that gas mixtures can be separated by means of gas separation membranes because of different permeabilities of the individual gases. In one way, to produce such gas separation membranes, polymers are processed to give hollow fibers or flat membranes. The membranes are characterized by a very thin separation layer so that the permeance of the membrane is as large as possible.

WO 2012/00727; WO 2013/098024; WO 2014/075850; KR10-1327337; KR10-1327338; U.S. pat. No. 6,565,626 B1; U.S. Pat. No. 6,168,649 B1; JP 2009-242773 A; each disclose membrane separation processes with three membrane separation stages, wherein a retentate stream from stage 3 and a permeate stream from stages 2 are recycled to the crude gas stream. WO 2012/00727; WO 2013/098024 and WO 2014/075850 represent the most optimized of all of these processes. In said patents an apparatus and a process are described which are optimized in view of product purity in combination with the lowest energy consumption. In other words, these processes provide two high pure product streams in an energy optimized way.

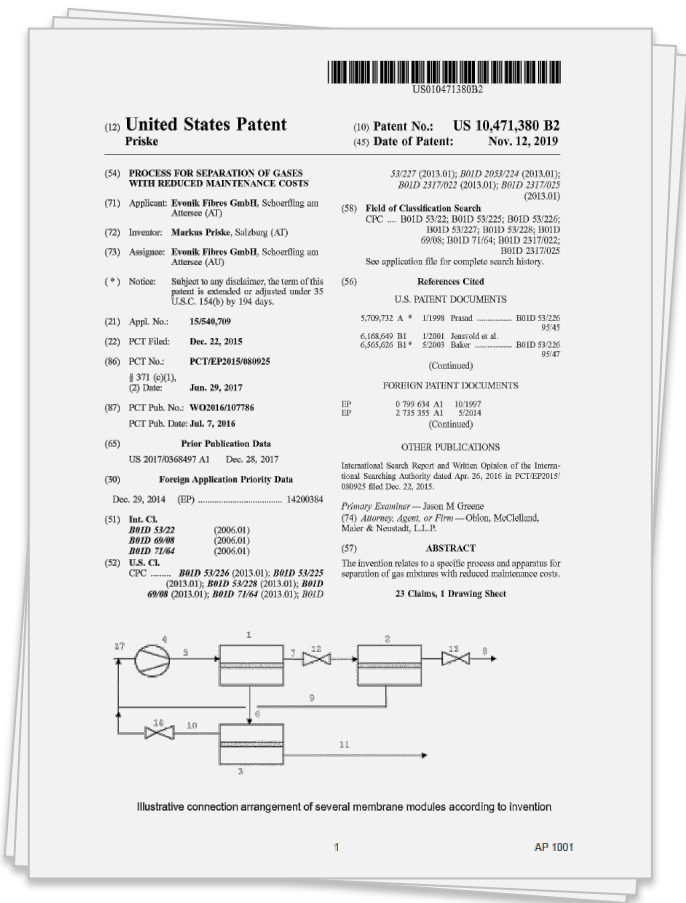
Priske-'380 | Recognizing an Unmet Need in Low-Cost Energy Environments



these processes. In said patents an apparatus and a process are described which are optimized in view of product purity in combination with the lowest energy consumption. In other words, these processes provide two high pure product streams in an energy optimized way.

In recent times, however, a new problem has occurred, that is not solved in a sufficient manner by the apparatuses and processes of the prior art. The problem is that some gas separation plants are operated at sites where energy is very cheap. At such locations the focus of operators of separation plants lies on investment and maintenance costs rather than on an energy saving. No sufficient solution for this problem has been found in the prior art.

Priske-'380 | Discovery of a Maintenance Cost Optimum

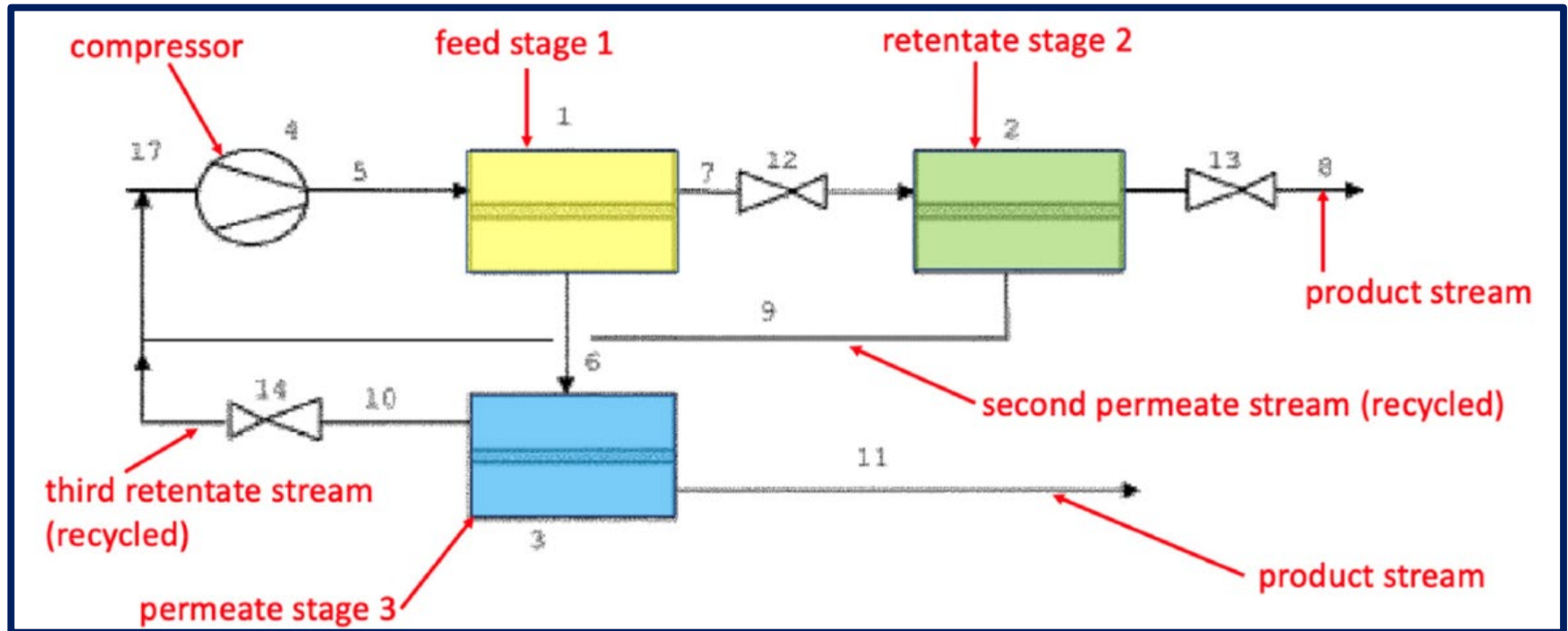


The inventors of the present invention surprisingly found, that beside of the energy optimum, as disclosed in WO 2012/00727, a maintenance cost optimum for three-stage membrane separation exists. This can be reached by increasing the total gas volume recycled in sum with the second permeate stream (9) and the third retentate stream (10), in combination with an increase of the total membrane capacity of the retentate separation stage (2) compared to the total membrane capacities of separation stage (1) and preferably also of stage (3), and a specific quotient of the pressure ratios of the permeate separation stage (3) over the feed stream separation stage (1).

The examples below show the invest and maintenance cost improvement over WO 2012/00727.

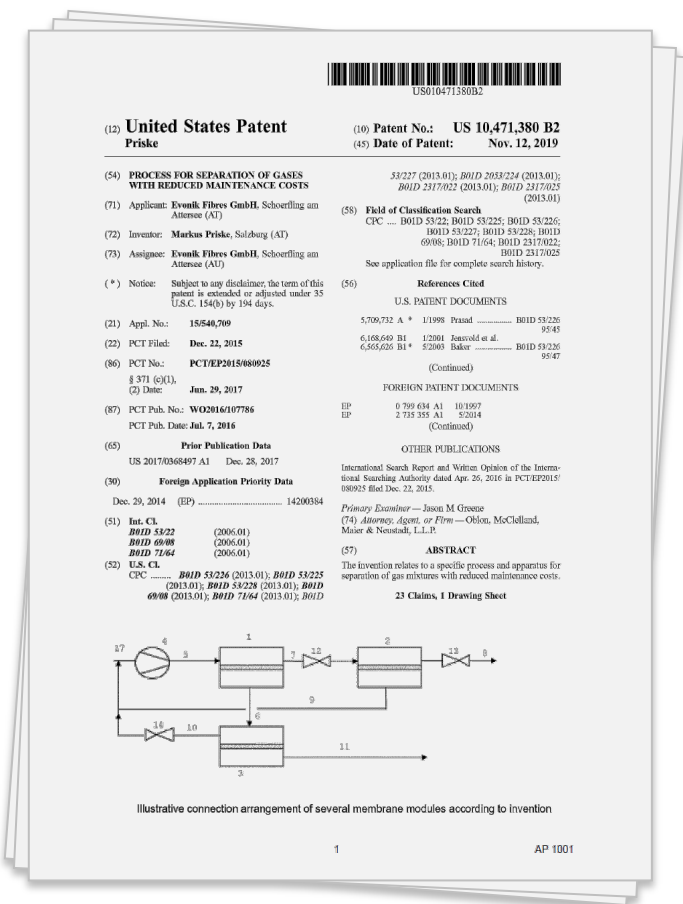
Ex. 1001, '380 Patent, 2:27-40

Figure 1 of the '380 Patent



Ex. 1001, '380 Patent, Fig. 1 (as annotated at Corrected Petition, 6)

Reference Example 1 of the '380 Patent



Reference Example 1

The Comparative-Example of WO 2012/000727 A1 was reproduced. Characteristics of this example are:

A recycled gas volume in sum of 86.7%

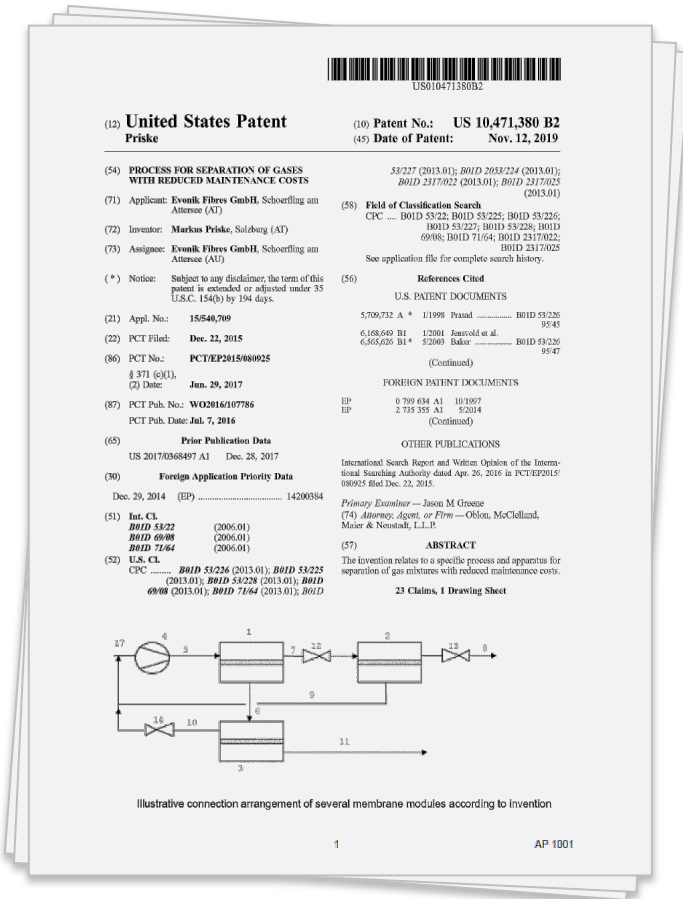
Identical membrane capacities in all three separation stages

A quotient of the pressure ratio over the permeate separation stage (3) to the pressure ratio over the feed stream separation stage (1) of 1.0.

Membrane selectivity for $\text{CO}_2/\text{CH}_4=20$ in all separation stages

Thus, the reference example differs from the present invention in that the membrane capacities are identical in all three stages.

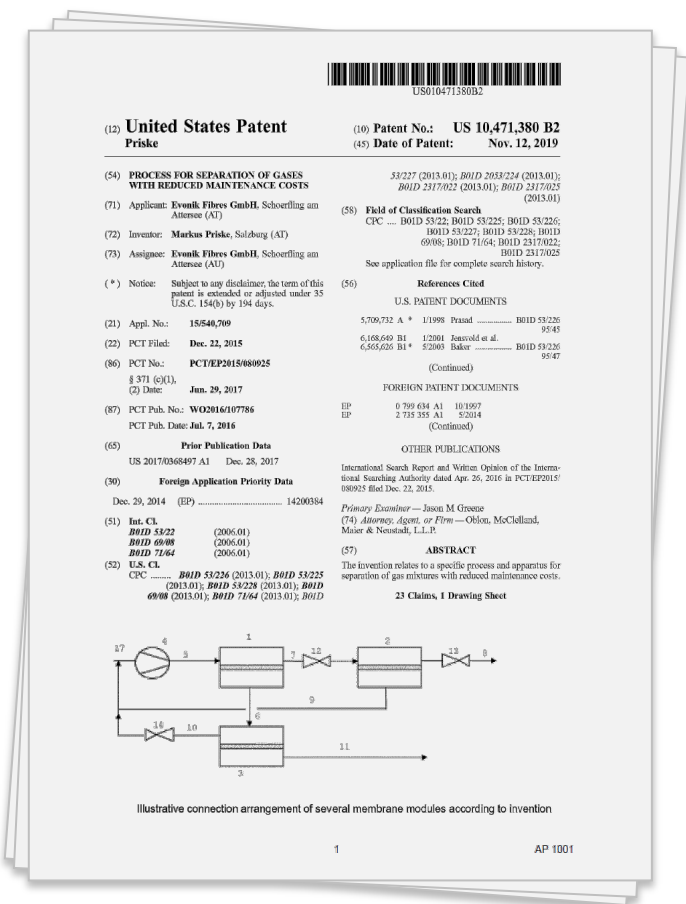
'380 Patent Disclosed Benefits of Claimed Invention



It has now surprisingly been found that the inventive process described herein and the inventive apparatus described herein can afford pure streams of permeate and retentate without requiring more than one compressor respectively without further purification of the permeate or retentate stream by other methods. The inventive apparatus thus enables simultaneous achievement of permeate and retentate streams in high purity. As shown in the examples, the new process, compared to the prior art processes, in particular WO 2012/00727, can be operated with a significantly reduced overall membrane capacity to obtain similar purification results. Thus, the costs for the membranes, which are expendable goods and have to be exchanged time by time, were reduced significantly.

Ex. 1001, '380 Patent, 2:13-26

Reference Example 1 is the Same as CE1 of Ungerank



Reference Example 1

The Comparative-Example of WO 2012/000727 A1 was reproduced. Characteristics of this example are:

A recycled gas volume in sum of 86.7%

Identical membrane capacities in all three separation stages

A quotient of the pressure ratio over the permeate separation stage (3) to the pressure ratio over the feed stream separation stage (1) of 1.0.

Membrane selectivity for $\text{CO}_2/\text{CH}_4=20$ in all separation stages

Thus, the reference example differs from the present invention in that the membrane capacities are identical in all three stages.

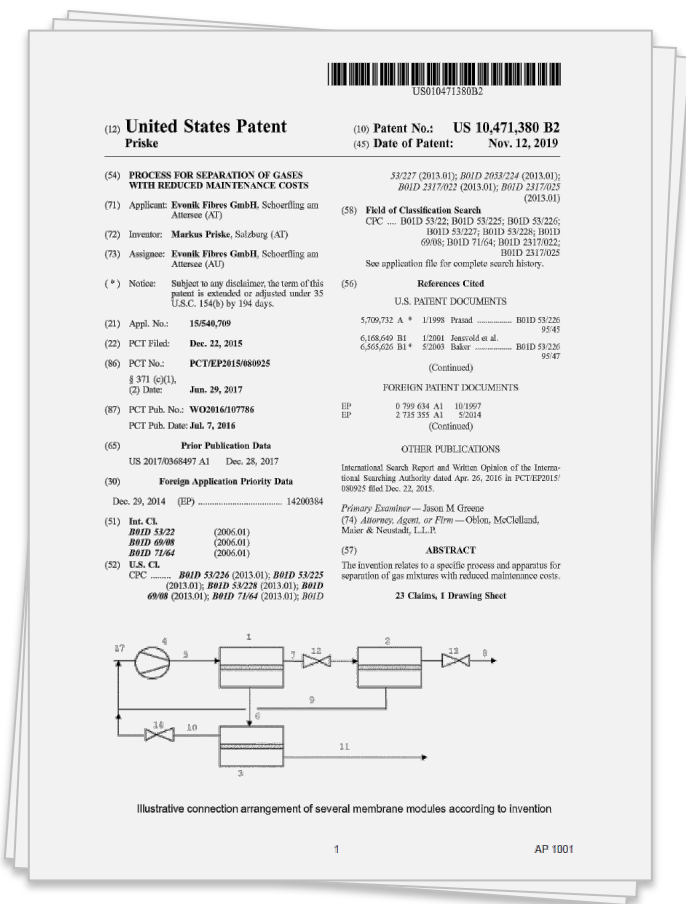
Ex. 1001, '380 Patent, 16:21-25
Ex. 2018, Baker May Dep., 29:21-25
Sur-Reply, 8

Example 1.1 Shows an Embodiment of the '380 Patent

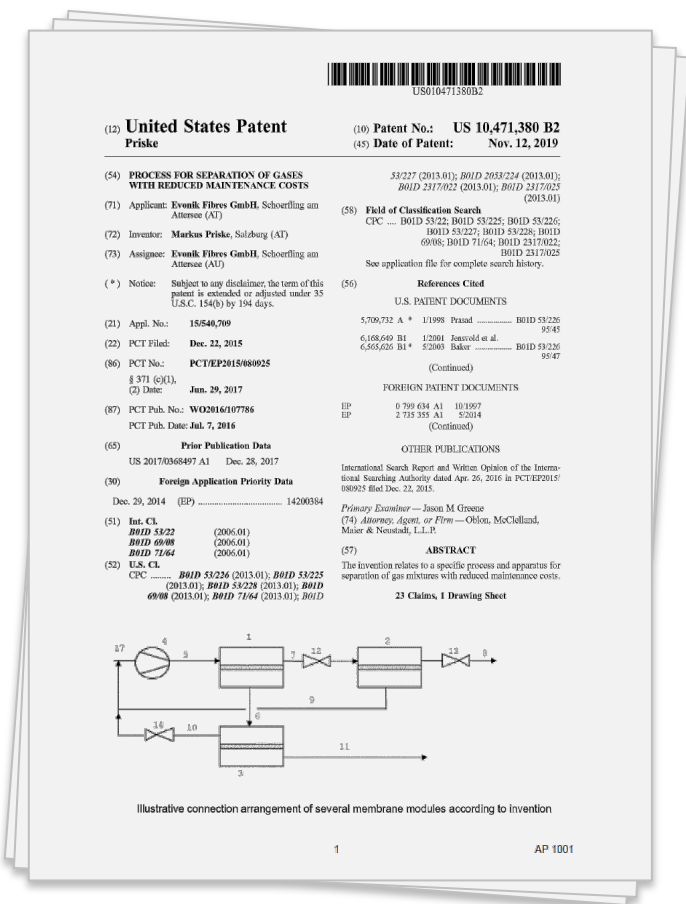
Example 1.1

Reference Example 1 was adopted in an inventive manner, i.e. the “recycling rate”, the “membrane capacity” of the retentate separation stage (2) and the “quotient of the pressure ratios of separation stage (3) over separation stage (1)” were increased. The recycling rate and the purities of product streams (8) and (11), however, were chosen identical respectively nearly identical to

As mentioned above, Example 1.1 differs from Reference example 1 in that the “recycling rate”, the “ratio of membrane capacity of stage (2) to stage (1) respectively of stage (2) to stage (3)” and the “quotient of the pressure ratio” were increased. The effect was, that the total membrane capacity, in sum for all three separation stages, of Example 1.1 was 14.9% lower than in Reference Example 1. In other words, in the inventive set-up of the apparatus respectively process, invest cost could be reduced because the required total membrane capacity could be reduced.



Example 1.2 Shows Another Embodiment of the '380 Patent



Example 1.2

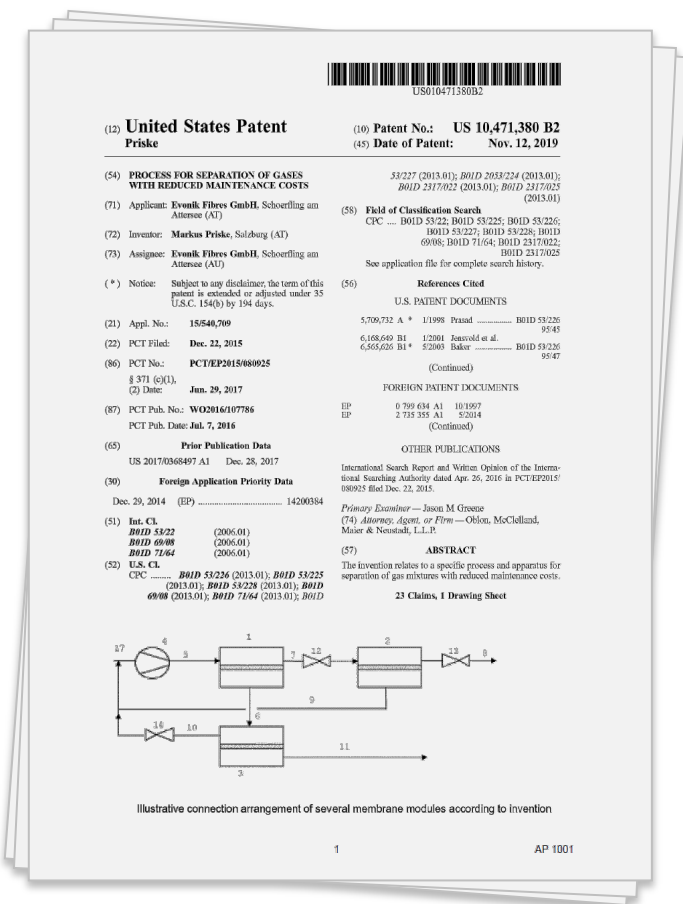
Reference Example 1 was reproduced again, however, in Example 1.2 only the parameters , “ratio of membrane capacity of stage (2) to stage (1) respectively of stage (2) to stage (3)” and the “quotient of the pressure ratio” were increased. In contrast to Example 1.1, the “recycling rate” was chosen identical to Reference Example 1 in order to show, what happens, if only two parameters were modified in an inventive manner.

The process parameters of Example 1.2 are summarized in Table 1 and 2 below:

In Example 1.2 a total saving of 14.5% of membrane capacity, in sum for all three separation stages, compared to Reference Example 1, was achieved.

This shows that the increase of the membrane capacity of separation stage (2) has a significant impact on the reduction of the total membrane capacity needed. Compared to Example 1.1, Example 1.2 shows, that an additional reduction of the overall required membrane capacity could be achieved if the recycling rate is increased, too.

Example 2 Shows Another Embodiment of the '380 Patent



Example 2

Reference Example 2 was modified in that the “recycling rate” was increased to 71% and the “membrane capacity of stage (2)” as well as the “quotient of pressure ratios” were increased, too. The operating temperature in all separation stages was 20° C. Details of the process parameters are given in Tables 5 and 6 below:

Due to the inventive modifications, the total membrane capacity required in Example 2 could be reduced, compared to Reference Example 2, by 15%.

Examples 1.1 and 1.2, with a membrane selectivity for CO₂/CH₄ of 20, and Example 2 with a selectivity for CO₂/CH₄ of 45 confirm, that the inventive apparatus and process set-up leads to improvements of the required overall membrane capacity for low as well as for higher selective membranes. In Example 5 below it will be demonstrated, that even membranes with a selectivity of 70 for CO₂/CH₄ can be used in the process of the invention. In all cases reduced invest and maintenance costs were achieved compared to a non-inventive three-stage separation process.

Ex. 1001, '380 Patent, 19:20-20:5
POR, 16-17

Examples 4-10 Show Additional Inventive Features of the '380 Patent



US 10,471,380 B2

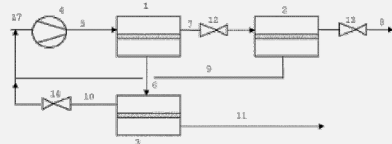
(12) United States Patent
Priske

(10) Patent No.: US 10,471,380 B2
(45) Date of Patent: Nov. 12, 2019

(54) PROCESS FOR SEPARATION OF GASES
WITH REDUCED MAINTENANCE COSTS

53/227 (2013.01); B01D 2053/224 (2013.01);
B01D 2112/023 (2013.01); B01D 2112/025

In Examples 1 to 10 different parameters were changed compared and directly compared to corresponding comparative examples. It could be shown, that process and apparatus of the invention are applicable for a wide range of operating very lucrative for applications in regions where energy is cheap and invest respectively maintenance costs are decisive.



Illustrative connection arrangement of several membrane modules according to invention

1

AP 1001

The total membrane capacity required in Example 4 was 12.4% below that of Reference Example 4. Accordingly, the process and apparatus of the invention is beneficial also for gas mixtures with a high methane content.

The total membrane capacity in Example 5 was 8.4% below that of Reference Example 5. This shows, that the process and apparatus of the invention are beneficial when using high selective membranes.

The total membrane capacity in Example 6 was 27.9% below that of Reference Example 6. This shows that process and apparatus of the invention are beneficial also, if low operating pressures are applied.

The total membrane capacity in Example 7 was 18.6% below that of Comparative Example 7. Thus, the process and apparatus of the invention are applicable at high operating pressures.

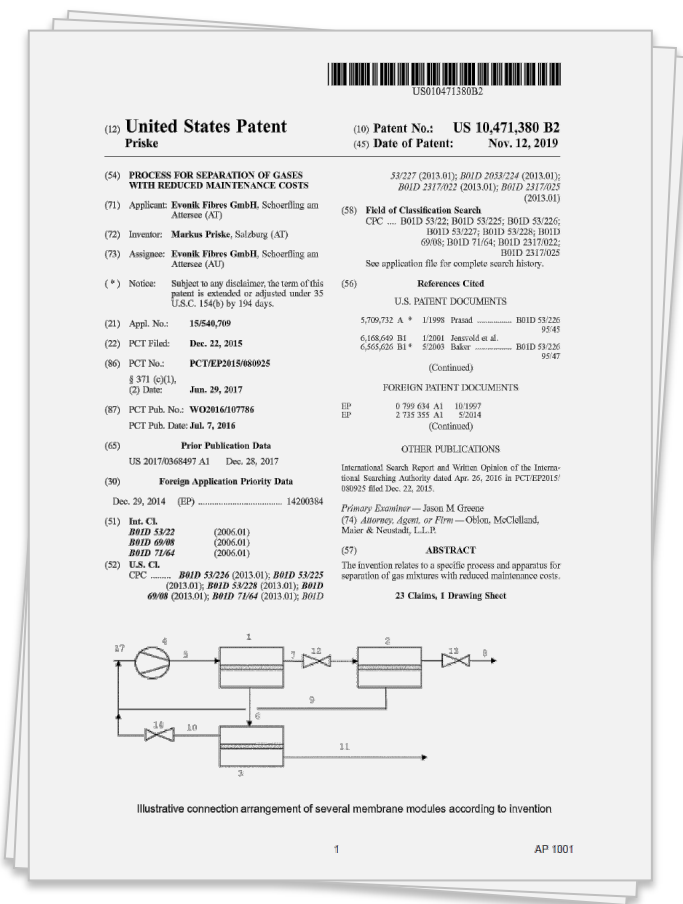
The total membrane capacity in Example 8 was 21.1% below that of Comparative Example 8. Thus, even if the “membrane capacity of stage (2)” is selected in an inventive manner but the “recycling rate” is not, a significant increase of the total membrane capacity has to be accepted.

This shows the synergistic effects of choosing the “membrane capacities of stage (2)” and of the “recycling rate” both in an inventive manner.

The total membrane capacity in Example 9 was 19.4% below that of Comparative Example 9 and that of Example 10 was 9.4% below that of Comparative Example 9. This shows that process and apparatus of the invention are also beneficial if more complex gas mixtures have to be separated.

Ex. 1001, '380 Patent, 27:61-28:59
POR, 16-17

Examples 11 and 12 Show Embodiments With Different Selectivities



Examples 11 and 12

In Example 11 membranes with different selectivities were used in different separation stages. In Example 12 the process parameters are very similar to Example 11, however, membranes with identical selectivities were used in all three stages. In both examples an apparatus according to FIG. 1 was used. The parameters of the simulations are given in Table 15 below:

Comparison of Example 11 and Example 12 reveals that the inventive process and apparatus is applicable with membranes with different selectivities in different separation stages but also with membranes with identical selectivities in all separation stages.

Priske-'380 | Relevant Dependent Claims

8. The method of claim 7, wherein the ratio of the total capacity of the gas separation membranes in the permeate separation stage to the total capacity of the gas separation membranes in the feed stream separation stage is in a range of from 0.8 to 1.

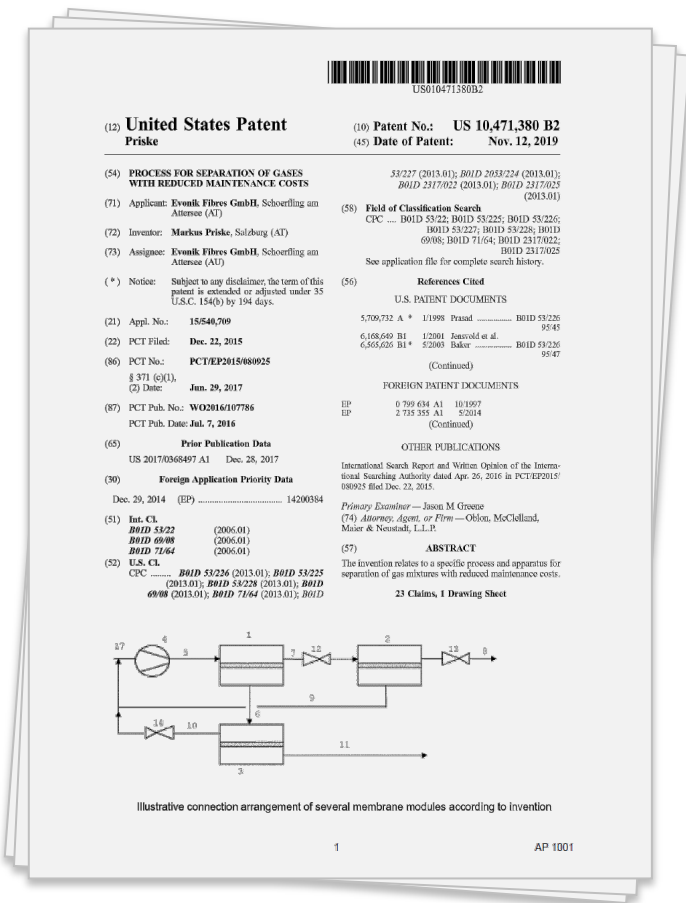
12. The method of claim 11, wherein the gas separation membranes in the feed stream separation stage have a pure gas selectivity for CO_2/CH_4 of from 20 to 100.

9. The method of claim 1, wherein the combined gas volume recycled with the second permeate stream and the third retentate stream is from 65 to 85% of the volume of the crude gas stream.

17. The method of claim 2, wherein the compressor is a multistage compressor and the second permeate stream, the third retentate stream, or both are introduced into the compressor between two compression stages.

10. The method of claim 1, wherein the quotient of the pressure ratio over the permeate separation stage to the pressure ratio over the feed stream separation stage is in a range of from 1 to 3.

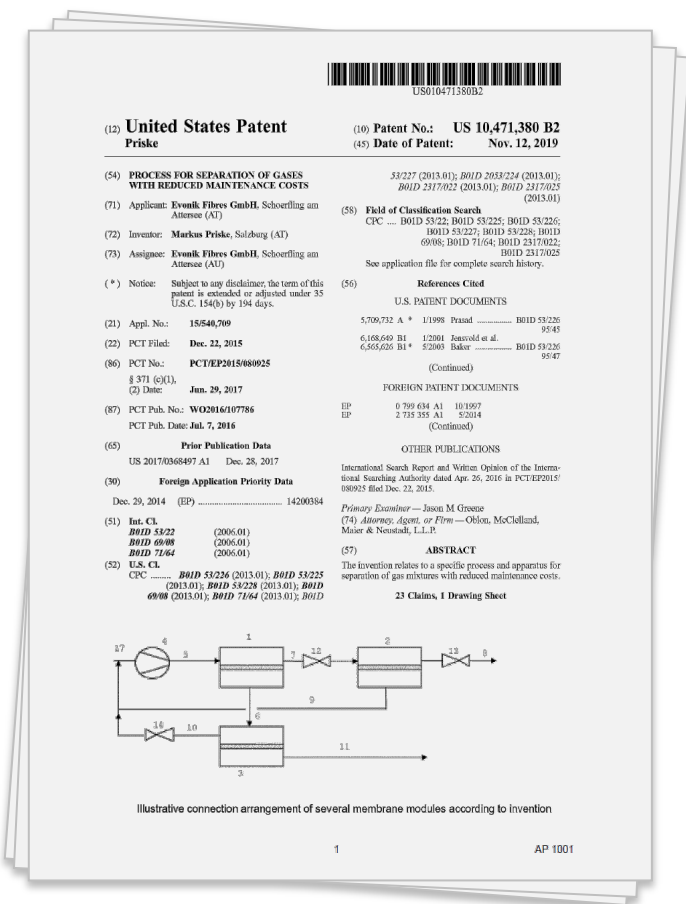
Priske-'380 Defines Membrane Capacity



Membrane capacity as used in the present invention is defined as the product of the membrane surface and the permeance of the membrane at operating temperature that is determined for nitrogen (Grade 4.8) under standard conditions. Standard conditions are described in detail in section measurement methods below. Membrane surface respectively membrane area is the external surface of the membrane as macroscopically visible. For its determination it is assumed that the membrane does not have pores and the external surface is homogeneous and smooth. In other words, the membrane surface of a flat membrane calculates as the product of length and width of the flat sheet and the membrane surface of hollow fiber membranes calculates as the product of length and external circumference of the hollow fiber.

Ex. 1001, '380 Patent, 2:53-67

Priske-'380 Defines Membrane Capacity



Membrane Capacity

The calculation method as well as the reference gas and the standard conditions are described below.

Hollow Fiber Membranes:

The membrane capacity of the hollow fiber membrane (HFM) calculates as follows:

$$\text{Capacity}_{(HFM)} = \text{Permeance}_{(HFM)} * \text{Surface}_{(HFM)}$$

Wherein the permeance(HFM) is tested under the following standard conditions:

Reference gas: Nitrogen, Grade 4.8

Temperature: instead of room temperature as applied above for the permeance measurement to calculate the selectivities; for the capacity calculation, the permeance is determined at the average operating temperature of the membrane, which is usually the operating temperature of the separation stage

Retentate pressure: 11 bara

Permeate pressure: 1.1 bara

and the surface(HFM) correlates to the external membrane surface as defined in the definitions section in this disclosure.

Asserted Prior Art

Asserted Prior Art | The Ungerank Patent

(12) **United States Patent**
Ungerank et al.

(10) **Patent No.:** **US 8,999,038 B2**
(45) **Date of Patent:** **Apr. 7, 2015**

(54) **PROCESS FOR SEPARATION OF GASES**
3/104 (2013.01); B01D 2256/24 (2013.01); B01D 2257/504 (2013.01); Y02C 10/10 (2013.01)

(75) **Inventors:** **Markus Ungerank**, Perg (AT); **Goetz Baumgarten**, Haltern am See (DE); **Markus Priske**, Dueseldorf (DE); **Harald Roegl**, Wallern an der Trattnach (AT)

(58) **Field of Classification Search**
USPC 95/45, 47, 51, 55; 96/4, 7, 9, 10, 14
See application file for complete search history.

(56) **References Cited**
U.S. PATENT DOCUMENTS
5,591,250 A 1/1997 Stern et al.
5,919,285 A 7/1999 Li et al.
6,168,649 B1 1/2001 Jensvold et al.
6,180,008 B1 1/2001 White
6,425,267 B1 * 7/2002 Baker et al. 95/47
6,565,626 B1 * 5/2003 Baker et al. 95/51
6,648,944 B1 * 11/2003 Baker et al. 95/51
2004/0168570 A1 * 9/2004 Franek 95/50
(Continued)

FOREIGN PATENT DOCUMENTS
JP 2009-242773 A 10/2009
WO 00/06293 2/2000
WO 2011/009919 1/2011

OTHER PUBLICATIONS
Franz Borgmann, "Modellierung und Optimierung von Gaspermeationsanlagen", Diplomaufgabe, Jun. 20, 1990, 206 pages (with English-language translation).
(Continued)

(73) **Assignee:** **Evonik Fibres GmbH**, Schörfing am Attersee (AT)

(*) **Notice:** Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 0 days.

(21) **Appl. No.:** **13/807,831**

(22) **PCT Filed:** **May 26, 2011**

(86) **PCT No.:** **PCT/EP2011/058636**
§ 371 (c)(1),
(2), (4) **Date:** **Dec. 31, 2012**

(87) **PCT Pub. No.:** **WO2012/000727**
PCT Pub. Date: **Jan. 5, 2012**

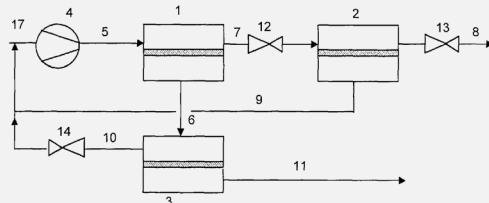
(65) **Prior Publication Data**
US 2013/0098242 A1 Apr. 25, 2013

(30) **Foreign Application Priority Data**
Jul. 1, 2010 (EP) 10168121

(51) **Int. Cl.**
B01D 53/22 (2006.01)
C10L 3/10 (2006.01)

(52) **U.S. Cl.**
CPC **B01D 53/225** (2013.01); **B01D 53/226** (2013.01); **B01D 53/227** (2013.01); **C10L**

58 Claims, 5 Drawing Sheets



Illustrative connection arrangement of several membrane modules according to invention

Patent No.: 8,999,038

Inventors: Ungerank, Baumgarten, Priske, Roegl

Assignee: Evonik Operations GMBH

Issue Date: Apr. 7, 2015

Asserted Prior Art | Ungerank Patent Teaches Multiple Configurations

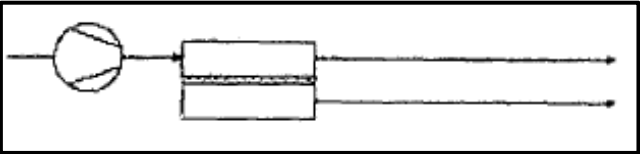


Fig. 2

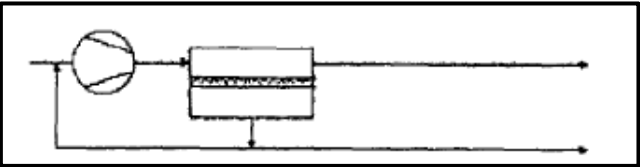


Fig. 3

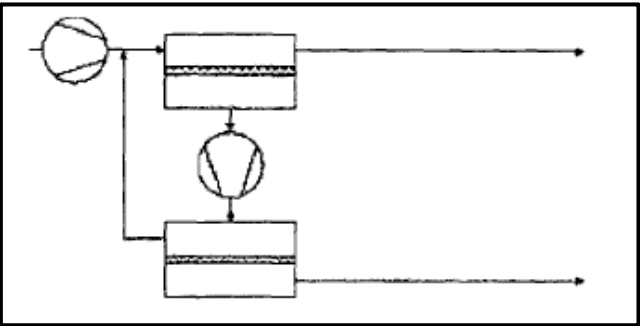


Fig. 4

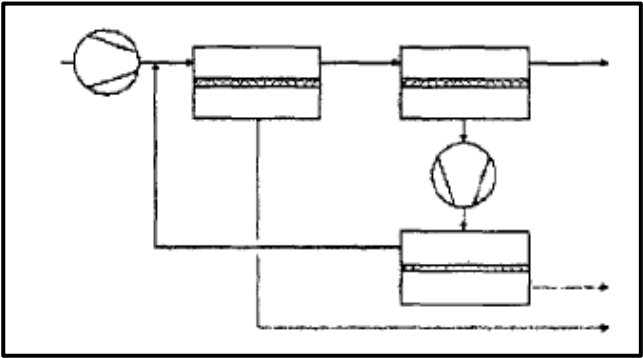


Fig. 5

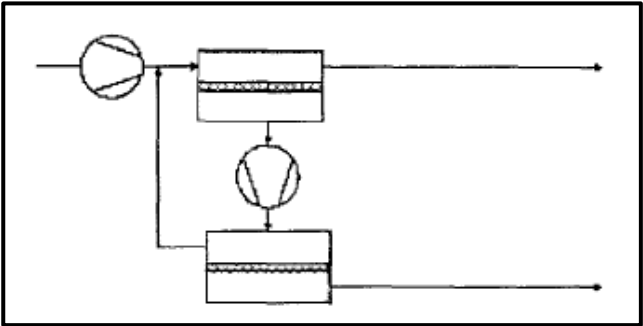


Fig. 6

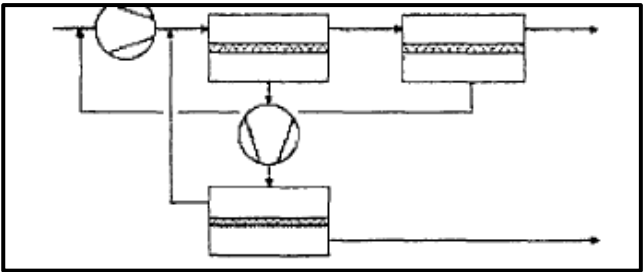


Fig. 7

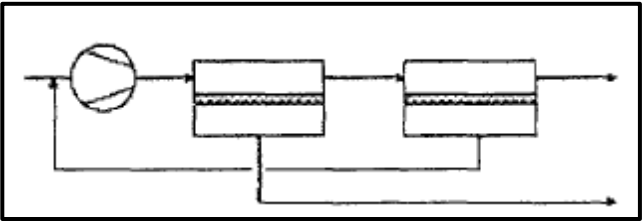


Fig. 8

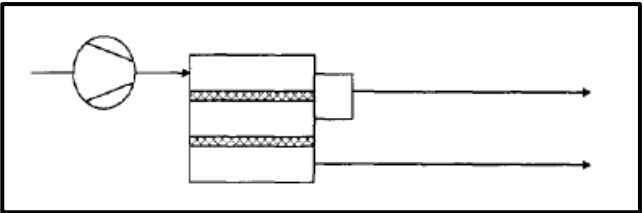


Fig. 9

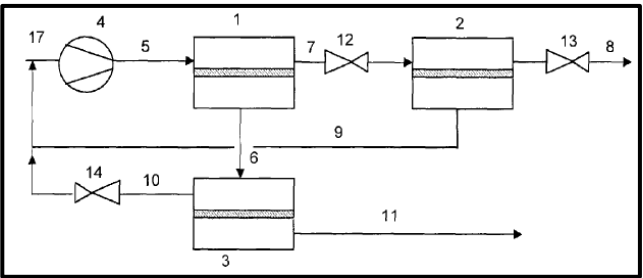


Fig. 11

Asserted Prior Art | Ungerank Patent Teaches Higher Selectivity

According to the invention, the gas separation membrane module (=ratio of permeate to retentate flow rate) is at least 30, even more preferably at least 40, and most preferably of at least 50. This results in a significant economic advantage, especially in the case of use of a one-stage compressor (4), it is necessary to compress less gas twice, which entails economic advantages in the operation of the plant. In the case of very selective membrane modules having a selectivity of 45, only approx. 35% of the gas introduced into the feed stream separation stage (1) as crude gas need be compressed twice; with a membrane module having a selectivity of only 10, it may be the case that the double compression is up to 300%. The figures of 35% and 300% are based on experiments in which a gas mixture with equimolar amounts of components A and B (=feed) was applied, with 98.5% component B present in the retentate gas of stage (2) and 99% of component B in the permeate stream of stage (3).

Membranes of higher selectivity have the advantage that the separation becomes more effective and less permeate has to be recycled from retentate separation stage (2), or less retentate from permeate separation stage (3).

Asserted Prior Art | Ungerank Patent Comparative Example 1

Ungerank Patent Comparative Example 1 is the same as Reference Example 1 in Priske-'380

COMPARATIVE EXAMPLE 1

Separation of a Mixture of Methane and Carbon Dioxide with a Mixing Ratio of 50 to 50 with a Moderately Selective Polyimide Membrane

The FIG. 11 shown connection arrangement was used. Each stage consisted of a hollow fiber membrane module consisting of hollow polyimide fibers from UBE (NM B01 A type). 1.78 m³/h of a crude gas mixture of 50% methane and 50% carbon dioxide, which corresponds approximately to a gas mixture of biogas, is introduced into a mixing chamber (not shown in FIG. 11) and then compressed to 25 bar together with recycled gas composed of gas streams (9) and (10). The gas which has been compressed and cooled to 20° C. is applied to the feed stream separation stage (1). The retentate of the feed stream separation stage (1) is subsequently passed via the first retentate stream (7) to the retentate separation stage (2). The permeate of retentate separation stage (2) has a volume flow rate of 0.743 m³/h with a methane content of 34.5% and a carbon dioxide content of 65.5%, and is recycled via the second permeate stream (9) into the mixing chamber and compressed again by the compressor (4).

The permeate of the feed stream separation stage (1) has a volume flow rate of 1.67 m³/h with a carbon dioxide content of 91.0% and a methane content of 9.0%, and is passed via the first permeate stream (6) as feed into the permeate separation stage (3). The pressure drop over the membrane of stage (1) does not extend to ambient pressure, but is limited to 4.2 bara by a reducing valve (14) on the retentate side of the permeate separation stage (3). This gives a third permeate stream (11) from permeate separation stage (3) of 0.885 m³/h with a composition of 99.0% carbon dioxide and only 1.0% methane. The third retentate stream (10) from the permeate separation stage (3) is 0.801 m³/h with a composition of 17.9% methane and 82.1% carbon dioxide and is recycled into the mixing chamber and compressed again. The sum of recycled gas streams (9) and (10) is therefore 1.544 m³/h or 86.7% based on the amount of gas to be separated supplied. Pure product streams are obtained with a relatively high double

The membranes used exhibit a moderate mixed gas selectivity for carbon dioxide over methane of 20.

Ex. 1006, Ungerank Patent, 10:16-67
Ex. 2018, Baker May Dep., 29:21-25
Sur-Reply, 8

Asserted Prior Art | Ungerank Patent Comparative Example 1

COMPARATIVE EXAMPLE 1

Separation of a Mixture of Methane and Carbon Dioxide with a Mixing Ratio of 50 to 50 with a Moderately Selective Polyimide Membrane

The FIG. 11 shown connection arrangement was used. Each stage consisted of a hollow fiber membrane module consisting of hollow polyimide fibers from UBE (NM B01 A type). 1.78 m³/h of a crude gas mixture of 50% methane and 50% carbon dioxide, which corresponds approximately to a gas mixture of biogas, is introduced into a mixing chamber (not shown in FIG. 11) and then compressed to 25 bar together with recycled gas composed of gas streams (9) and (10). The gas which has been compressed and cooled to 20° C. is applied to the feed stream separation stage (1). The retentate of the feed stream separation stage (1) is subsequently passed via the first retentate stream (7) to the retentate separation stage (2). A reducing valve (13) on the retentate side of the retentate separation stage (2) is set to 18.2 bara and thus determines the driving force through the membrane of membrane separation stages (1) and (2). The retentate of the retentate separation stage (2) has a content of 98.5% methane and 1.5% carbon dioxide. 0.895 m³/h of this mixture leaves retentate separation stage (2). The permeate of retentate separation stage (2) has a volume flow rate of 0.743 m³/h with a methane content of 34.5% and a carbon dioxide content of 65.5%, and is recycled via the second permeate stream (9) into the mixing chamber and compressed again by the compressor (4).

The FIG. 11 shown connection arrangement was used. Each stage consisted of a hollow fiber membrane module consisting of hollow polyimide fibers from UBE (NM B01 A type). 1.78 m³/h of a crude gas mixture of 50% methane and

The permeate of the feed stream separation stage (1) has a volume flow rate of 1.67 m³/h with a carbon dioxide content of 91.0% and a methane content of 9.0%, and is passed via the first permeate stream (6) as feed into the permeate separation stage (3). The pressure drop over the membrane of stage (1) does not extend to ambient pressure, but is limited to 4.2 bara by a reducing valve (14) on the retentate side of the permeate separation stage (3). This gives a third permeate stream (11) from permeate separation stage (3) of 0.885 m³/h with a composition of 99.0% carbon dioxide and only 1.0% methane. The third retentate stream (10) from the permeate separation stage (3) is 0.801 m³/h with a composition of 17.9% methane and 82.1% carbon dioxide and is recycled into the mixing chamber and compressed again. The sum of recycled gas streams (9) and (10) is therefore 1.544 m³/h or 86.7% based on the amount of gas to be separated supplied. Pure product streams are obtained with a relatively high double compression rate. The membranes used exhibit a moderate mixed gas selectivity for carbon dioxide over methane of 20.

Asserted Prior Art | Ungerank Patent Example 1 is More Economically Favorable than Comparative Example 1

EXAMPLE 1

Separation of a mixture of methane and carbon dioxide with highly selective membranes

The FIG. 11 shows a process for separating a mixture of methane and carbon dioxide. Each stage consists of high pressure separation area of polyimide fibers. Austrian patent with a precipitation

The membranes used exhibit a high mixed gas selectivity for carbon dioxide over methane of 45. The high selectivity of 45 compared to 20 in the comparative example therefore enables a more economically favorable process by a lower recompression of 31.2% rather than 86.7%.

C. 1 m³/h of a crude gas mixture of 50% methane and 50% carbon dioxide, which corresponds approximately to a gas mixture of biogas, is introduced into a mixing chamber and then compressed to 25 bar together with recycled gas composed of gas streams (9) and (10). The gas which has been compressed and cooled to 20° C. is applied to the feed stream separation stage (1). The retentate of this stage is passed via the first retentate stream (7) to the retentate separation stage (2). A reducing valve (13) on the retentate side of the retentate separation stage (2) is set to 18.4 bara and thus determines the driving force through the membrane of membrane separation stages (1) and (2). The retentate of the retentate separation

Asserted Prior Art | Ungerank Patent Example 2 Teaches That It is Desirable to Minimize Methane Loss Below 0.5%

EXAMPLE 2

Separation of a Mixture of Methane Dioxide with a Mixing Ratio of 50 Highly Selective Polyimide Memb Methane Content in the Permeate Stream than 0.5%

In order to satisfy various legislators, minimize the methane loss into the atmosphere, the shown connection arrangement was used to lower the concentration of methane in the third permeate stream below 0.5% by volume. Each stage consisted of a hollow fiber membrane module consisting of highly selective polyimide

The permeate of the feed stream separation stage (1) has a volume flow rate of $0.607 \text{ m}^3/\text{h}$ with a carbon dioxide content of 93.1% and a methane content of 6.9%, and is passed via the first permeate stream (6) as feed into the permeate separation stage (3). The pressure drop over the membrane of stage (1) does not extend to ambient pressure, but is limited to 4.4 bara by a reducing valve (14) on the retentate side of the permeate separation stage (3). This gives a third permeate stream (11) from permeate separation stage (3) of $0.495 \text{ m}^3/\text{h}$ with a composition of 99.5% carbon dioxide and only 0.5% methane. The third retentate stream (10) from the permeate separation stage (3) is $0.112 \text{ m}^3/\text{h}$ and has a composition of 35% methane and 65% carbon dioxide and is recycled into the mixing chamber and compressed again. The sum of recycled streams (9) and (10) is therefore $0.356 \text{ m}^3/\text{h}$ or 35.6% based on the amount of gas to be separated supplied. Pure product streams are obtained with a moderate double compression rate. The membranes used exhibit a high mixed gas selectivity for carbon dioxide over methane of 45.

In order to satisfy various legislators, it is necessary to minimize the methane loss into the atmosphere. The FIG. 11 shown connection arrangement was used to lower the concentration of methane in the third permeate stream (11) to below 0.5% by volume.

with a precipitation bath temperature of 40°C . rather than 10°C . $1 \text{ m}^3/\text{h}$ of a crude gas mixture of 50% methane and 50% carbon dioxide, which corresponds approximately to a gas mixture of biogas, is introduced into a mixing chamber and then compressed to 25 bar together with recycled gas composed of gas streams (9) and (10). The gas which has been compressed and cooled to 20°C . is applied to the feed stream separation stage (1). The retentate of this stage is passed via the first retentate stream (7) to the retentate separation stage (2). A reducing valve (13) on the retentate side of the retentate separation stage (2) is set to 18.1 bara and thus determines the driving force through the membrane of membrane separation stages (1) and (2). The retentate of the retentate separation stage (2) has a content of 98.5% methane and 1.5% carbon dioxide. $0.505 \text{ m}^3/\text{h}$ of this mixture leaves retentate separation stage (2). The permeate of retentate separation stage (2) has a volume flow rate of $0.244 \text{ m}^3/\text{h}$ with a methane content of 26.1% and a carbon dioxide content of 73.9%, and is recycled via the second permeate stream (9) into the mixing chamber and compressed again by the compressor (4).

*Ex. 1006, Ungerank Patent, Example 2
POR, 39*

Asserted Prior Art | Scholz



Structural optimization of membrane-based biogas upgrading processes

Marco Scholz, Michael Alders
RWTH Aachen University, Chemical Process Engineering

ARTICLE INFO

Article history:
Received 17 February 2014
Received in revised form 8 August 2014
Accepted 14 August 2014
Available online 28 September 2014

Keywords:
Process modeling
Structural optimization
Gas permeation
Biogas upgrading

1. Introduction

The depletion of fossil resources and the need for renewable energies, Biogas upgrading energy supply. Contrary to solar energy, biogas is produced continuously. Biogas will be a future energy supply as it is a renewable energy source. The separation of CH_4 and CO_2 is a well-known process. Biogas upgrading refers to the separation of CH_4 and CO_2 and purified so that it can be used as natural gas. Permeation membranes are well known for gas separation [2–4]. Their application in biogas upgrading has several advantages over conventional gas separation processes. Gas permeation operates with the low elevated pressure so that it can directly be integrated into the gas grid [1]. Secondly gas permeation process is simple, which is particularly advantageous for application [3].

The objective of the study at hand is the structural optimization of membrane-based biogas upgrading process by optimization. Here, (i) the optimal process configuration and (ii) the optimal required membrane areas in the various stages are determined.

*Corresponding author. Tel.: +49 241 80 95 95.
E-mail address: marco.scholz@rwth-aachen.de
<https://doi.org/10.1016/j.memsci.2014.08.032>
0276-5688/© 2014 Published by Elsevier B.V.

Structural optimization of membrane-based biogas upgrading processes

Marco Scholz, Michael Alders, Theresa Lohaus, Matthias Wessling*

RWTH Aachen University, Chemical Process Engineering, Turmstr.46, 52064 Aachen, Germany



ARTICLE INFO

Article history:

Received 17 February 2014
Received in revised form 8 August 2014
Accepted 14 August 2014
Available online 28 September 2014

Keywords:

Process modeling
Structural optimization
Gas permeation
Biogas upgrading

ABSTRACT

Gas permeation membranes are well known for separating CO_2 and CH_4 . Single stage processes cannot provide both, high product gas purity and high recovery at the same time, but multistage processes do. Commonly, the design of multistage gas permeation processes relies on heuristics and experience, so that often sub-optimal separation processes are designed. We apply a structural optimization approach to biogas upgrading processes. This systematic method determines the most profitable process layout including membrane areas and pressures required for two commercial membranes. Furthermore, we determine the optimal selectivity and CO_2 permeance following the upper bound of the Robeson plot. The process model is implemented using General Algebraic Modeling System (GAMS). For commercial membranes a three stage gas permeation process is the most profitable process which operates with only a single compressor. Optimizing the process layout together with the membrane properties a two stage process is the best process configuration. CO_2/CH_4 selectivities of approximately 120 are optimal. The impact of changed feed conditions and required product purity was investigated. The results highlight that for biogas upgrading commercial gas permeation membranes already provide powerful separation characteristics. The optimal membrane processes are capable to economically compete with conventional biogas upgrading techniques.

© 2014 Published by Elsevier B.V.

Asserted Prior Art | Scholz Makes Certain Economic Assumptions

Table 3
Economic parameters used for optimization of a membrane based biogas upgrading plant.

Costs	Unit	Value
Membrane module costs	$\frac{\text{Euro}}{\text{m}^2}$	55
Electrical costs	$\frac{\text{Euro}}{\text{kWh}}$	0.08
Commission of purified gas	$\frac{\text{Euro}}{\text{m}^3}$	0.2
Membrane lifetime	year	4
Annual operation	h	8,000
Interest rate	—	0.09
Amortization period	years	8



Contents list

Journal of

journal homepage:

Structural optimization of membrane upgrading processes

Marco Scholz, Michael Alders, Theresa Lohaus

KVTH Aachen University, Chemical Process Engineering, Turmstr.46, 52064 Aachen, Germany

ARTICLE INFO

Article history:
Received 19 February 2014
Received in revised form 8 August 2014
Accepted 18 August 2014
Available online 28 September 2014

Keywords:
Process modeling
Structural optimization
Gas permeation
Biogas upgrading

ABSTRACT

Gas permeation membranes provide both, high gas fluxes and high selectivity. Commonly, the design of a membrane-based biogas upgrading process is determined by the optimization of the membrane area. The process model for a membrane-based biogas upgrading process is the only single component. The impact of changes in the separation characteristics of conventional biogas

1. Introduction

The depletion of fossil resources triggers the transition to renewable energies. Biogas upgrading will contribute to the energy supply. Contrary to solar and wind power, biogas is produced continuously. Biogas will considerably contribute to the future energy supply as it is a renewable resource. Biogas upgrading refers to the separation of CH_4 and CO_2 , where a CH_4 rich gas is purified so that it can be used as natural gas substitute [1]. Permeation membranes are well known for separating CO_2 and CH_4 [2–4]. Their application in biogas upgrading offers several advantages over conventional gas separation techniques. First, gas permeation operates with the upgraded product gas below elevated pressure so that it can directly be injected into the natural gas grid [1]. Secondly, gas permeation modules are robust and the process is simple, which is particularly suitable for on-site application [5].

The objective of the study at hand is to design a membrane-based biogas upgrading process based on structural process optimization. Here, (i) the optimal process configuration, (ii) the required membrane areas in the various stages and (iii) the

from synthesis gas, and hydrogen recovery from refinery streams. They obtained new process designs which reduce the costs by 20% compared to process designs previously reported in the literature. They applied stochastic optimization using a simulated annealing algorithm.

Qi and Henson [9,10] report on CO_2/CH_4 separation in natural gas treatment and enhanced oil recovery. Here, only recycle compressors are investigated as the raw gas is already pressurized.

*Corresponding author. Tel.: +49 241 80 99470; fax: +49 241 80 92282.
E-mail address: marco.scholz@kvth-wt-achen.de (M. Scholz).

<https://doi.org/10.1016/j.memsci.2014.08.032>
0376-7388/\$ - 2014 Published by Elsevier B.V.

Asserted Prior Art | Scholz, Figure 5



Journal of
journal homepage: v

Structural optimization of membrane upgrading processes

Marco Scholz, Michael Alders, Theresa Lohaus,
RWTH Aachen University, Chemical Process Engineering, Turmstr. 46, 52064 Aachen

ARTICLE INFO

Article history:
Received 19 February 2014
Received in revised form 8 August 2014
Accepted 18 August 2014
Available online 28 September 2014

Keywords:
Process modeling
Structural optimization
Gas permeation
Biogas upgrading

ABSTRACT

Gas permeation membranes provide both, high permeability and selectivity, which are often sub-optimal for biogas upgrading. Including membrane modules in the process model to determine the optimal configuration of membranes is the only way to optimize a single compression stage process. The impact of changes in the separation characteristics of conventional biogas

1. Introduction

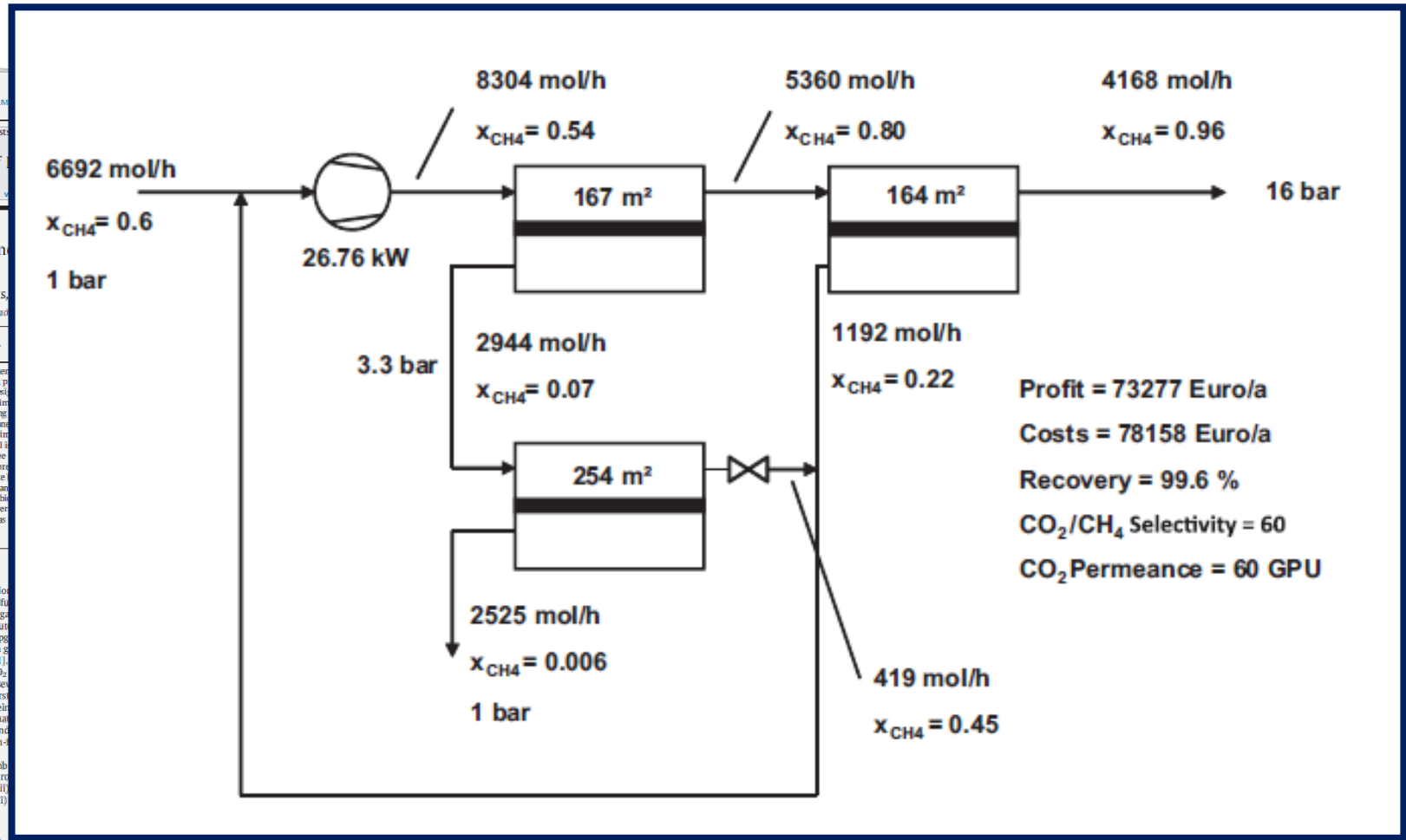
The depletion of fossil resources triggers the transition to renewable energies. Biogas upgrading will contribute to future energy supply. Contrary to solar and wind power, biogas is produced continuously. Biogas will considerably contribute to future energy supply as it is a renewable resource. Biogas upgrading refers to the separation of CH_4 and CO_2 , where a CH_4 rich gas is produced so that it can be used as natural gas substitute [1]. Permeation membranes are well known for separating CO_2 from CH_4 [2–4]. Their application in biogas upgrading offers several advantages over conventional gas separation techniques. First, gas permeation operates with the upgraded product gas being at elevated pressure so that it can directly be injected into the natural gas grid [1]. Secondly, gas permeation modules are robust and the process is simple, which is particularly suitable for on-site application [3].

The objective of the study at hand is to design a membrane-based biogas upgrading process based on structural process optimization. Here, (i) the optimal process configuration, (ii) the required membrane areas in the various stages and (iii) the

*Corresponding author. Tel.: +49 241 80 99470; fax: +49 241 80 92262.

E-mail address: marco.scholz@rwth-aachen.de (M. Scholz).

<https://doi.org/10.1016/j.memsci.2014.08.032>
0270-7388/© 2014 Published by Elsevier B.V.



compressors are investigated as the raw gas is already pressurized.

Asserted Prior Art | Scholz, Figure 7(a)



Journal of Membrane Science
journal homepage: www.elsevier.com/locate/jms

Structural optimization of membrane upgrading processes

Marco Scholz, Michael Alders, Theresa Lohaus
RWTH Aachen University, Chemical Process Engineering, Turmstr. 46, 52064 Aachen, Germany

ARTICLE INFO

Article history:
Received 19 February 2014
Received in revised form 8 August 2014
Accepted 18 August 2014
Available online 28 September 2014

Keywords:
Process modeling
Structural optimization
Gas permeation
Biogas upgrading

ABSTRACT

Gas permeation membranes provide both, high permeability and high selectivity. Commonly, the design of a membrane-based process is determined by the optimization of the membrane area. The process model includes a three-stage process. The impact of changes in the separation characteristics of conventional biogas

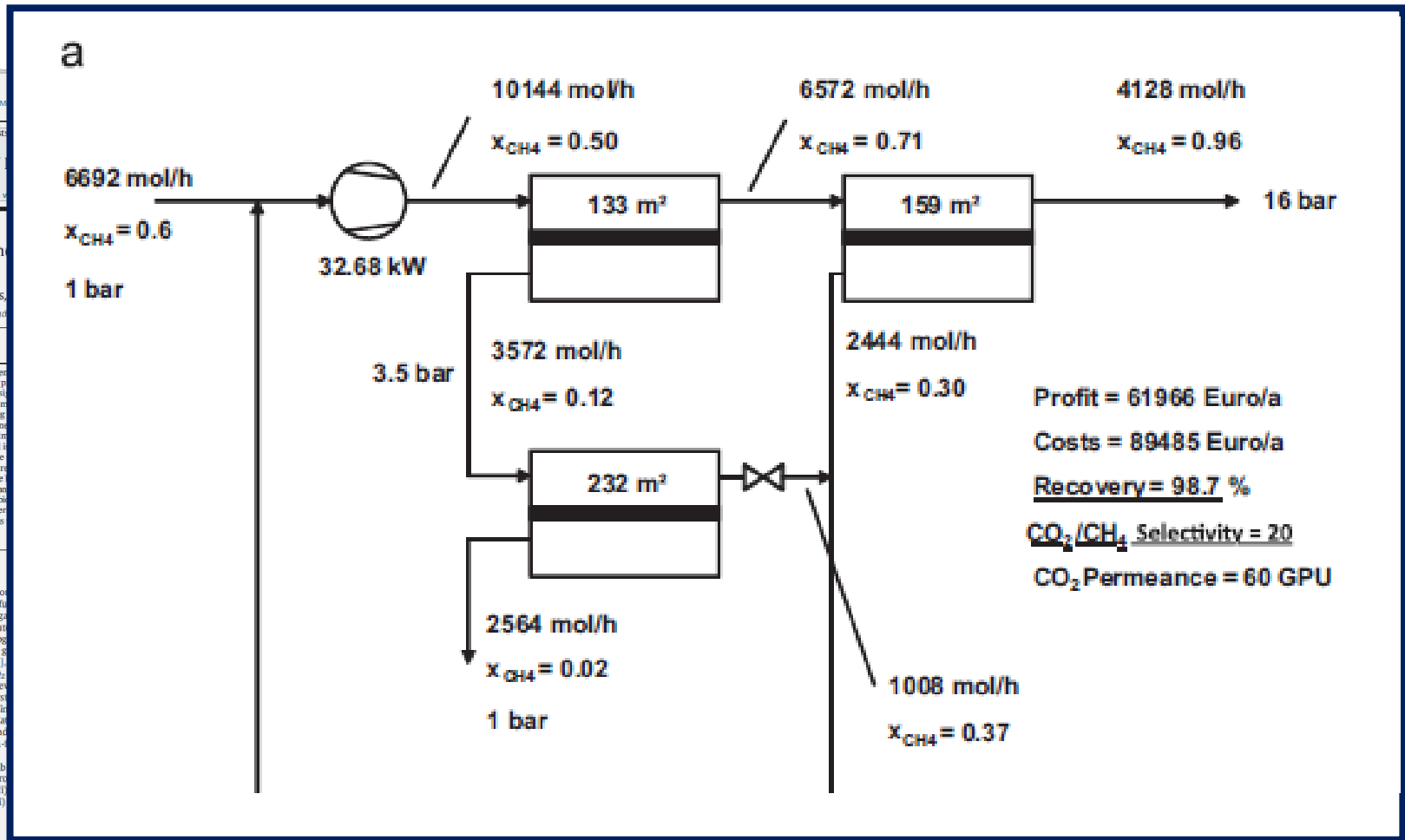
1. Introduction

The depletion of fossil resources triggers the transition to renewable energies. Biogas upgrading will contribute to future energy supply. Contrary to solar and wind power, biogas is produced continuously. Biogas will considerably contribute to future energy supply as it is a renewable resource. Biogas upgrading refers to the separation of CH_4 and CO_2 , where a CH_4 rich gas is produced so that it can be used as natural gas substitute [1]. Permeation membranes are well known for separating CO_2 from CH_4 [2–4]. Their application in biogas upgrading offers several advantages over conventional gas separation techniques. First, gas permeation operates with the upgraded product gas being elevated pressure so that it can directly be injected into the natural gas grid [1]. Secondly, gas permeation modules are robust and process is simple, which is particularly suitable for on-site application [3].

The objective of the study at hand is to design a membrane-based biogas upgrading process based on structural process optimization. Here, (i) the optimal process configuration, (ii) the required membrane areas in the various stages and (iii) the required

*Corresponding author. Tel.: +49 241 80 99470; fax: +49 241 80 92262.
E-mail address: marco.scholz@cpce.rwth-aachen.de (M. Scholz).

<https://doi.org/10.1016/j.memsci.2014.08.032>
0376-7388/\$ - 2014 Published by Elsevier B.V.



compressors are investigated as the raw gas is already pressurized.

Asserted Prior Art | Scholz, Figure 7(b)



Journal of
journal homepage: v

Structural optimization of membrane upgrading processes

Marco Scholz, Michael Alders, Theresa Lohaus,
RWTH Aachen University, Chemical Process Engineering, Turmstr. 46, 52064 Aachen

ARTICLE INFO

Article history:
Received 17 February 2014
Received in revised form
8 August 2014
Accepted 18 August 2014
Available online 28 September 2014

Keywords:
Process modeling
Structural optimization
Gas permeation
Biogas upgrading

ABSTRACT

Gas permeation membranes provide both, high permeability and high selectivity. Commonly, the design of a membrane-based process is often sub-optimal due to the lack of a process model that can determine the optimal membrane area. The process model is a three-stage process. The impact of changes in the separation characteristics of conventional biogas

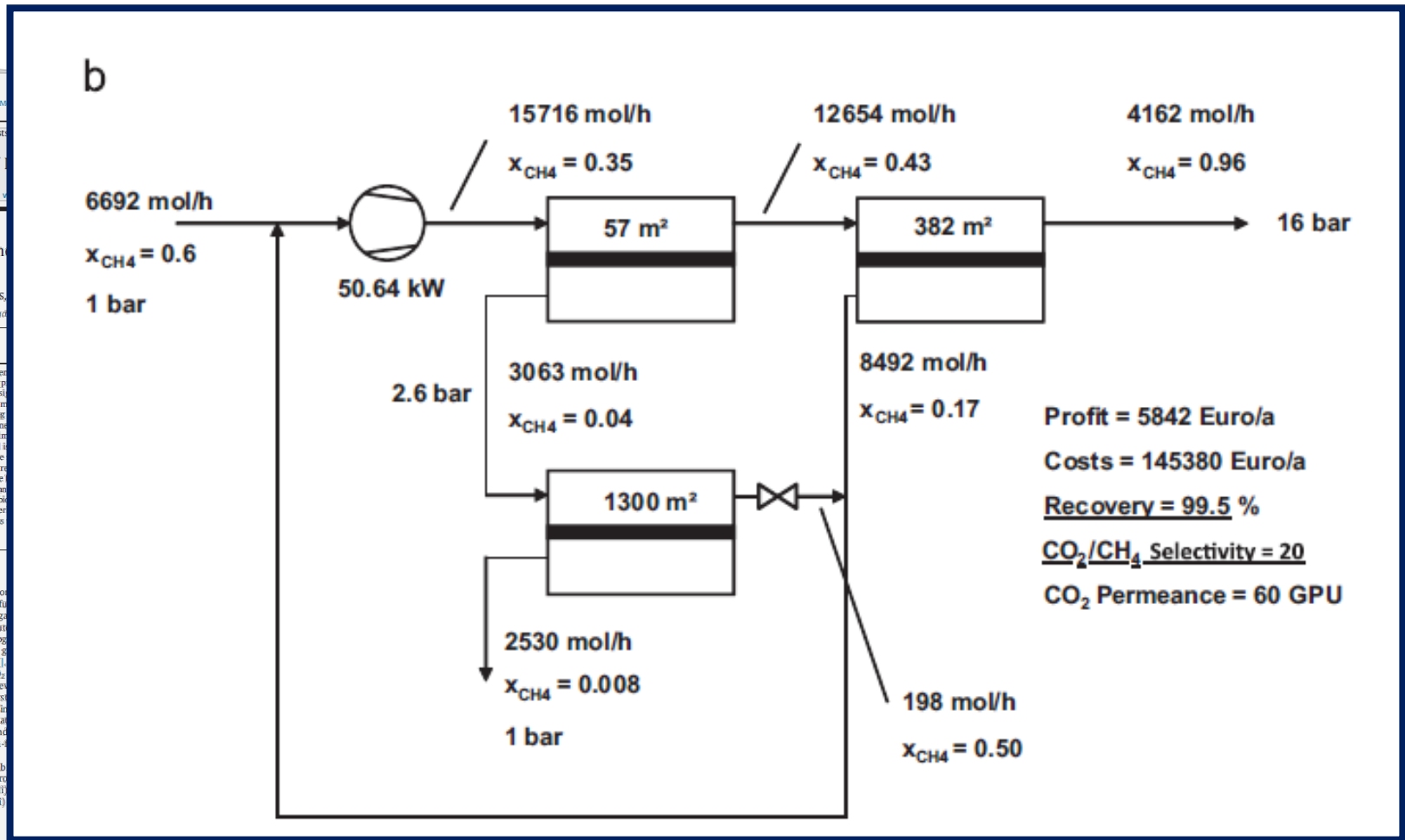
1. Introduction

The depletion of fossil resources triggers the transition to renewable energies. Biogas upgrading will contribute to future energy supply. Contrary to solar and wind power, biogas is produced continuously. Biogas will considerably contribute to future energy supply as it is a renewable resource. Biogas upgrading refers to the separation of CH_4 and CO_2 , where a CH_4 rich gas is produced so that it can be used as natural gas substitute [1]. Permeation membranes are well known for separating CO_2 from CH_4 [2–4]. Their application in biogas upgrading offers several advantages over conventional gas separation techniques. First, gas permeation operates with the upgraded product gas being at elevated pressure so that it can directly be injected into the natural gas grid [1]. Secondly, gas permeation modules are robust and process is simple, which is particularly suitable for on-site application [3].

The objective of the study at hand is to design a membrane-based biogas upgrading process based on structural process optimization. Here, (i) the optimal process configuration, (ii) the required membrane areas in the various stages and (iii) the

*Corresponding author. Tel.: +49 241 80 99470; fax: +49 241 80 92252.
E-mail address: marco.scholz@cpce.rwth-aachen.de (M. Scholz).

<https://doi.org/10.1016/j.memsci.2014.08.032>
0376-7388/© 2014 Published by Elsevier B.V.



compressors are investigated as the raw gas is already pressurized.

Overview of the Grounds

Overview of the Asserted Grounds

Ground	Claims Challenged	35 U.S.C. §	Reference(s)
I	1-5, 7, 9, 11-23	103	Ungerank Patent, Scholz
II	1-13, 16, 17, 21, 23	103	Scholz

Disputed Issues

Key Deficiencies of the Petition

- Ground I Is Statutorily Defective
 - The Ungerank Patent Is Not Prior Art
 - Assertion of the Ungerank Application First Identified in the Corrected Petition Would Have Been Time-Barred
- Petitioner's Original Obviousness Theory Under Ground I is Legally Flawed and Technically Deficient
- Petitioner's New Obviousness Theory Under Ground I in Reply is Untimely and Unsupported
- Ground I Also Fails Because Petitioner Has Not Demonstrated Motivation to Combine
- Ground II Fails Because Petitioner's Obviousness Theory Is Incorrect, Including Under *Titanium Metals*

Ground I

The Ungerank Patent is Not Prior Art

The Original Petition Relied on § 102(a)(2)

U.S. Patent No. 8,999,038 (“Ungerank”) is prior art under §102(a)(2).

Ungerank issued under §151, names another inventor than the '380 patent's named inventor, and was filed May 26, 2011, effectively before the earliest effective filing date of the '380 patent. AP-1006, 1.

The Ungerank Patent Is Not Prior Art Under 102(a)(2)

102(b) EXCEPTIONS.—

(2) DISCLOSURES APPEARING IN APPLICATIONS AND PATENTS.—A disclosure shall not be prior art to a claimed invention under subsection (a)(2) if—

(C) The subject matter disclosed and the claimed invention, not later than the effective filing date of the claimed invention, were owned by the same person or subject to an obligation of assignment to the same person.

(12) **United States Patent**
Ungerank et al.

(73) Assignee: **Evonik Fibres GmbH**, Schörfling am
Attersee (AT)

Ex. 1006

(12) **United States Patent**
Priske

(73) Assignee: **Evonik Fibres GmbH**, Schoerfling am
Attersee (AU)

Ex. 1001

The Corrected Petition Purports to Rely on 35 U.S.C. § 102(a)(1)

The portions of U.S. Patent No. 8,999,038 (AP-1006), cited by column and line numbers herein and in AP-1003, are prior art under § 102(a)(1), as they were published on April 25, 2013 in U.S. Patent App. Pub. No. 2013/0098242 (AP-1038) (“Ungerank”). The publication’s specification and figures are identical to the patent. *Compare* AP-1006 to AP-1038. Ungerank issued under § 151, names another inventor than the ’380 patent’s named inventor, and was filed May 26, 2011, effectively before the earliest effective filing date of the ’380 patent. AP-1006, 1.

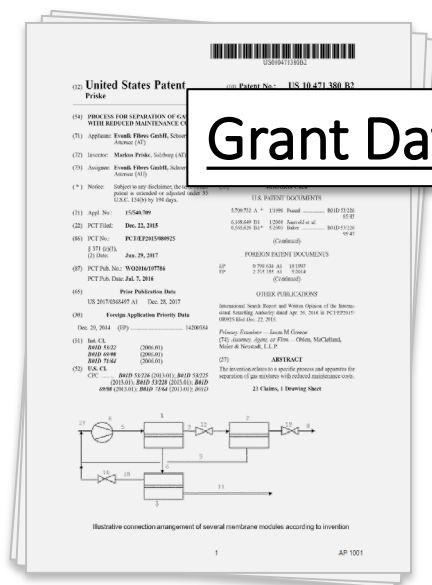
The Ungerank Patent Is Not Prior Art Under 102(a)(1)

MPEP § 2152.02(a)

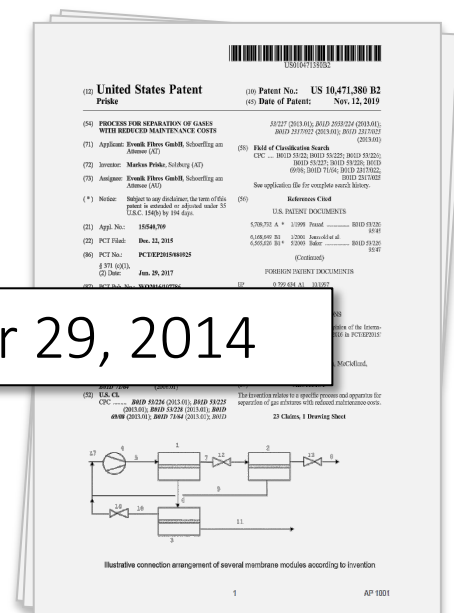
The effective date of the patent for the purposes of determining whether the patent qualifies as prior art under AIA 35 U.S.C. §102(a)(1) is the grant date of the patent.

Grant Date of Ungerank Patent: April 7, 2015

Priority Date of Priske-380: December 29, 2014



Ex. 1006, Ungerank Patent



Ex. 1001, '380 Patent

Ground I Was Instituted Based On the Ungerank Patent

a. Ungerank

Ungerank is a U.S. patent titled “Process for Separation of Gas,” filed May 26, 2011, and issued April 7, 2015, to EvonikFibres GmbH. Ex. 1006, codes (22), (45), (54), (73). The lead inventor is Markus Ungerank. *Id.*, code (75). Markus Priske, the sole inventor of the ’380 Patent, is also named as an inventor of Ungerank. *Id.*, code (75); Ex. 1001, code (72). Figure 11 of Ungerank is identical to Figure 1 of the ’380 Patent. *Compare* Ex. 1006, Fig. 11, *with* Ex. 1001, Fig. 1.

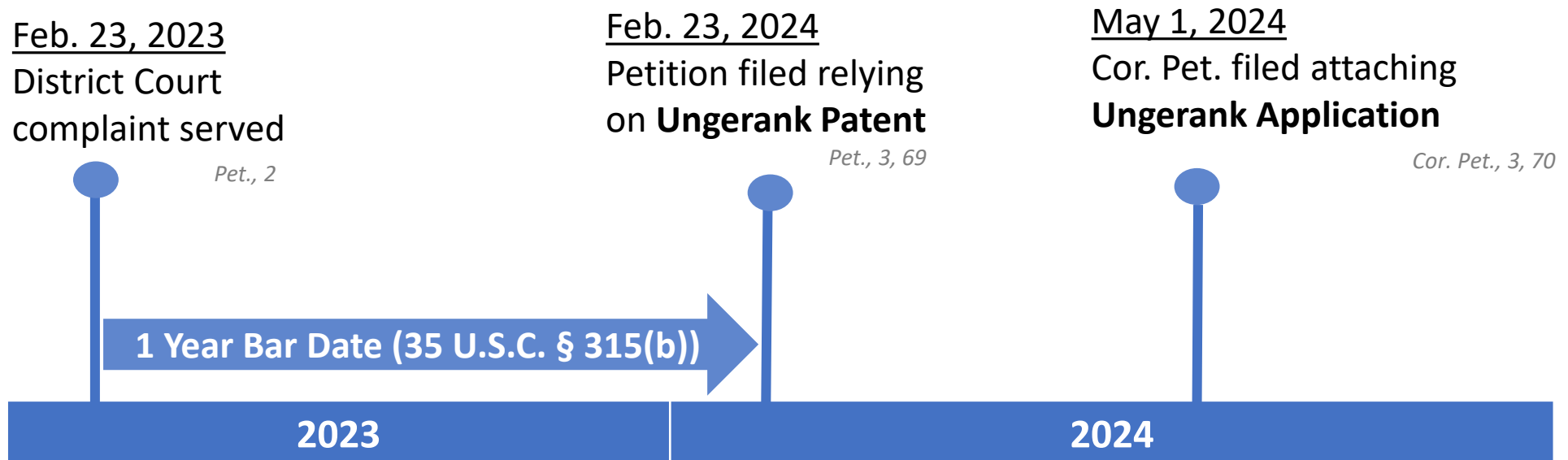
Decision Granting Institution, 7

The ’380 Patent makes reference to the Ungerank³ patent, relied on by Petitioner in support of Ground 1.

³ U.S. Patent No. 8,999,038, published Jan. 5, 2012 (Ex. 1006).

Decision Granting Institution, 3

Assertion of the Ungerank Application Would Have Been Time-Barred



35 U.S.C. § 315(b)

An inter partes review may not be instituted if the petition requesting the proceeding is filed more than 1 year after the date on which the petitioner, real party in interest, or privy of the petitioner is served with a complaint alleging infringement of the patent.

Issue Not Waivable

5 U.S.C. § 706

“The reviewing court shall ...

(2) hold unlawful and set aside agency action ... found to be ...

(C) in excess of statutory ... authority, or limitations ...”

SAS Inst., Inc. v. Iancu

“[T]he Director exceeded his statutory authority by limiting the review to fewer than all of the claims SAS challenged.”

584 U.S. 357, 371 (2018)

Samsung Elecs. Am., Inc. v. Prisia Eng'g Corp.

“It would not be proper for the Board to cancel claims on a ground that is unavailable in an IPR.”

948 F.3d 1342, 1353 (Fed. Cir. 2020)

Petitioner's Original Obviousness Theory Is Incorrect

Petitioner's Original Theory Regarding UCE1

Because Ungerank's Comparative Example 1 does not provide details on the membranes' areas (or their ratios), a POSITA would have found it obvious to combine Ungerank's teachings on three-stage systems with lower-selectivity membranes with Scholz's teachings in Figure 7 on optimized surface area ratios (proportional to the capacity ratios). AP-1003, ¶¶119-142. In doing so, a POSITA would have arrived at the Challenged Claims. *Id.*

Corrected Petition, 19

Petitioner's Original Theory Regarding UCE1

C. Ground 1 – Claims 1-5, 7, 9, 11-23 are obvious over Ungerank in view of Scholz

As described above and addressed further below, Ungerank's Comparative Example 1 discloses every limitation in claim 1 except for element [1f.ii]. AP-1003, ¶¶119-195. Element [1f.ii] requires that the capacity of the membranes in the retentate separation stage be higher than the capacity of the membranes in the feed stream separation stage. In other words, the capacity *ratio* must be greater than 1. *Id.*, ¶119.

Corrected Petition, 18

Because Ungerank's Comparative Example 1 does not provide details on the membranes' areas (or their ratios), a POSITA would have found it obvious to combine Ungerank's teachings on three-stage systems with lower-selectivity membranes with Scholz's teachings in Figure 7 on optimized surface area ratios (proportional to the capacity ratios). AP-1003, ¶¶119-142. In doing so, a POSITA would have arrived at the Challenged Claims. *Id.*

Corrected Petition, 19

111. Ungerank's Comparative Example 1 does not describe the membranes' areas, which as I discuss below, is related to their capacities.

Ex. 1003, Baker Decl., ¶ 111

134. However, Ungerank's Comparative Example 1 does not provide any details on the membranes' surface areas. A POSITA would therefore have sought guidance elsewhere on membranes' surface areas (and their ratios) in three-stage, one-compressor systems with relatively low-selectivity membranes.

Ex. 1003, Baker Decl., ¶ 134

153. As I discussed above, a POSITA would have been motivated to combine the information in Scholz's Figure 7 regarding surface area ratios (and therefore by extension, capacity ratios) for optimized three-stage systems with low-selectivity membranes with Ungerank's teachings on three-stage systems using lower-selectivity membranes, since Ungerank does not provide details on the membranes' areas in Comparative Example 1.

Ex. 1003, Baker Decl., ¶ 153

Petitioner's Original Theory Regarding UCE1

Petitioner further argues that, because Ungerank's Comparative Example 1 is silent as to membrane surface area, one would look to Scholz for guidance as to membrane size and size ratios. *Id.* at 19, 23; Ex. 1003 ¶¶ 134–136.

Decision Granting Institution, 13

Based on the present record, we agree with Petitioner's contention that one seeking to design a system employing lower-cost, lower-selectivity membranes would have looked to Comparative Example 1 of Ungerank which employs relatively lower-selectivity membranes (selectivity of 20) and achieves "[p]ure product streams . . . with a relatively high double compression rate." Ex. 1006, 10:64–66; Pet. 22. As Ungerank's Comparative Example 1 is silent regarding membrane surface area, a designer of a system employing lower-cost, lower-selectivity membranes would have reason to look to Scholz for guidance regarding membrane area.

Decision Granting Institution, 19

Petitioner's Original Theory Is Factually Incorrect: The Membrane Area Ratios of CE1 are 1:1

Because Ungerank's Comparative Example 1 does not provide details on the membranes' areas (or their ratios), a POSITA would have found it obvious to combine Ungerank's teachings on three-stage systems with lower-selectivity membranes with Scholz's teachings in Figure 7 on optimized surface area ratios (proportional to the capacity ratios). AP-1003, ¶¶119-142. In doing so, a POSITA would have arrived at the Challenged Claims. *Id.*

Corrected Petition, 19

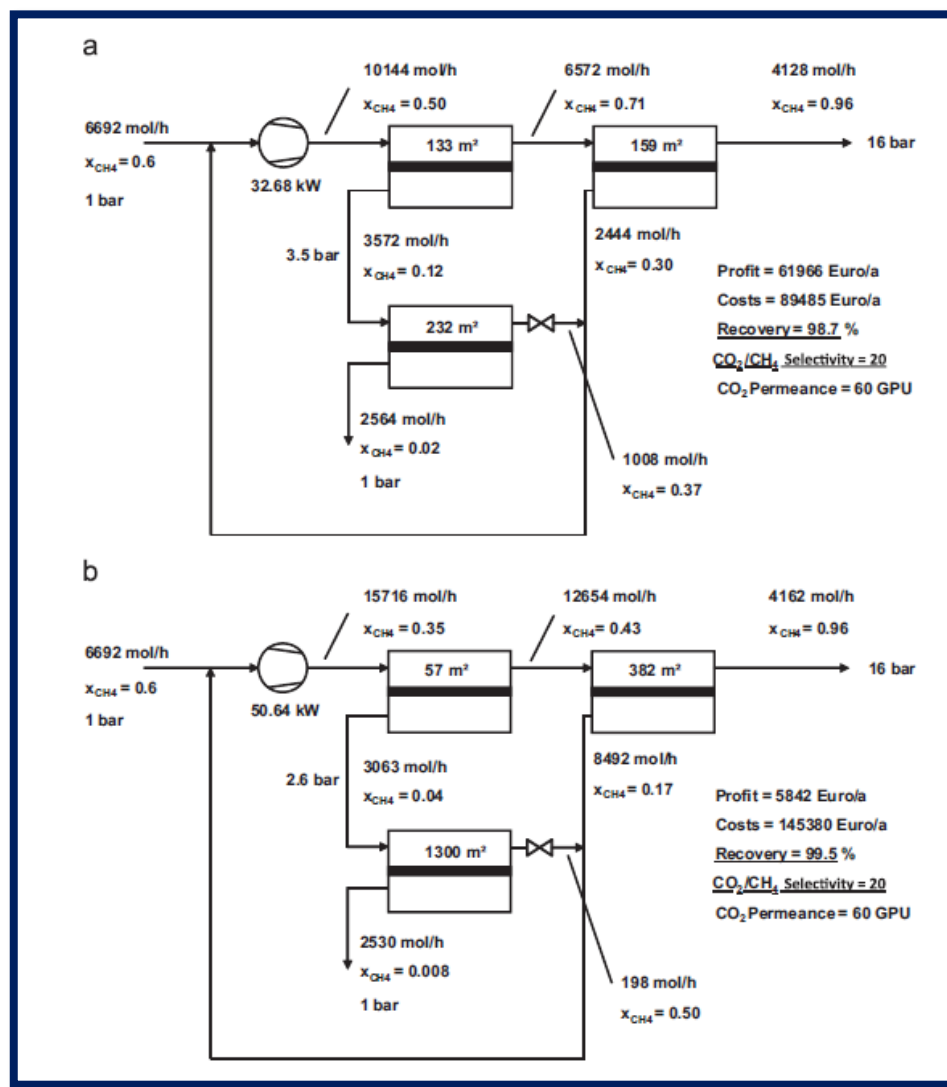
COMPARATIVE EXAMPLE 1

Separation of a Mixture of Methane and Carbon Dioxide with a Mixing Ratio of 50 to 50 with a Moderately Selective Polyimide Membrane

The FIG. 11 shown connection arrangement was used. Each stage consisted of a hollow fiber membrane module consisting of hollow polyimide fibers from UBE (NM B01 A type). 1.78 m³/h of a crude gas mixture of 50% methane and

Ex. 1006, Ungerank Patent, 10:16-27

Scholz Figures 7a and 7b Do Not Provide Capacity Values



Ex. 2007, Scholz, FIG. 7
POR 31

Petitioner and its Expert Improperly Assume Capacity Means The Equivalent of Membrane Area

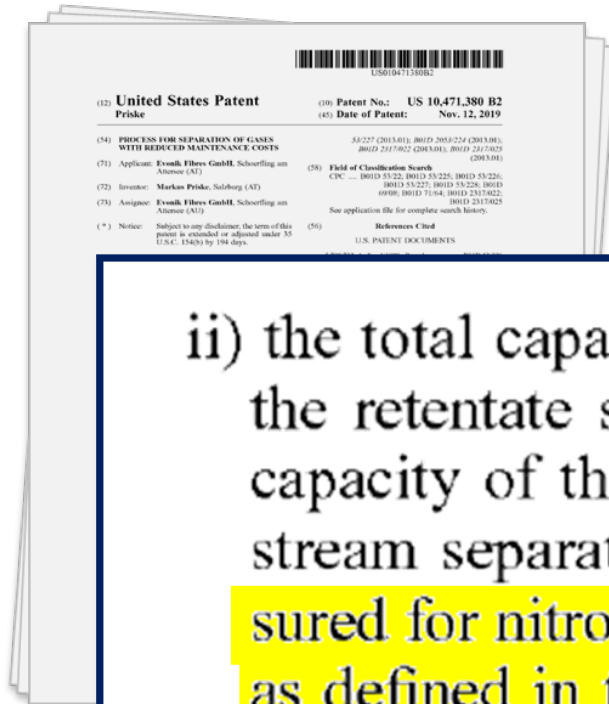


Q. And you said that, in your view, the patent defines that term "capacity" to mean membrane area; is that correct?

A. The equivalent of membrane area. It's proportional to membrane area. It doesn't tell you how many square meters, but it does tell you that the — when it's talking about the capacity of something is bigger than the capacity of the other thing, it means the membrane area of this one is bigger than the membrane area of the other.

*Ex. 2009, Baker Dec. Dep., 82:22-83:13
POR, 31*

Petitioner Fails to Address All Requirements of the Claim



The invention claimed is:

1. A method for separating gases from a crude gas stream comprising at least two components, wherein in an apparatus comprising a feed stream separation stage, a retentate separation stage and a permeate separation stage, each stage being a membrane separation stage with gas separation membranes,

- a feed stream is separated in the feed stream separation stage into a first permeate stream and a first retentate stream,
- the first retentate stream is separated in the retentate

ii) the total capacity of the gas separation membranes in the retentate separation stage is higher than the total capacity of the gas separation membranes in the feed stream separation stage, the total capacity being measured for nitrogen Grade 4.8 under standard conditions as defined in the description, and

capacity of the gas separation membranes in the feed stream separation stage, the total capacity being measured for nitrogen Grade 4.8 under standard conditions as defined in the description, and

- iii) the quotient of the pressure ratio over the permeate separation stage to the pressure ratio over the feed stream separation stage is in a range of from 0.5 to 8.

Ex. 1001, Priske-'380 Claim 1
POR, 31-32

Petitioner Fails to Apply Proper Construction for Capacity

Membrane capacity as used in the present invention is defined as the product of the membrane surface and the permeance of the membrane at operating temperature that is determined for nitrogen (Grade 4.8) under standard conditions. Standard conditions are described in detail in section measurement methods below. Membrane surface respectively membrane area is the external surface of the membrane as macroscopically visible. For its determination it is assumed that the membrane does not have pores and the external surface is homogeneous and smooth. In other words, the membrane surface of a flat membrane calculates as the product of length and width of the flat sheet and the membrane surface of hollow fiber membranes calculates as the product of length and external circumference of the hollow fiber.

Ex. 1001, '380 Patent, 2:15-67

Membrane Capacity

The calculation method as well as the reference gas and the standard conditions are described below.

Hollow Fiber Membranes:

The membrane capacity of the hollow fiber membrane (HFM) calculates as follows:

$$\text{Capacity}_{(HFM)} = \text{Permeance}_{(HFM)} * \text{Surface}_{(HFM)}$$

Wherein the permeance(HFM) is tested under the following standard conditions:

Reference gas: Nitrogen, Grade 4.8

Temperature: instead of room temperature as applied above for the permeance measurement to calculate the selectivities; for the capacity calculation, the permeance is determined at the average operating temperature of the membrane, which is usually the operating temperature of the separation stage

Retentate pressure: 11 bara

Permeate pressure: 1.1 bara

and the surface(HFM) correlates to the external membrane surface as defined in the definitions section in this disclosure.

Ex. 1001, '380 Patent, 14:51:15:5

Petitioner's Assumption is Circular and Lacks Merit



69. Dr. Baker's circular argument (at paragraphs 156-158)—that identical *CO₂ permeances* must mean the ratio of membrane capacities correlate to the ratio of membrane surface areas, and thus the ratio of membrane surface capacities *measured under nitrogen (Grade 4.8)* must correlate to the ratio of membrane surface areas—ignores the fact that the permeances of the different gases, e.g., CO₂ and nitrogen, and the ratios of those permeances may vary.

*Ex. 2008, Geise Decl., ¶169
POR, 32-33*

Dr. Baker does not Assert Error in Any Opinion Dr. Geise Offered



Q. So you agree that you reviewed Dr. Geise's declaration, but your supplemental declaration doesn't identify any errors that he made in that declaration, correct?

A. That's correct, I guess.



*Ex. 2018, Baker May Dep., 12:12-17
Sur-Reply, 23*

Permeance Depends on a Number of Factors



70. Even where two membranes are composed of cellulose acetate and have a permeance for CO₂ of 60 GPU, the permeance for nitrogen (Grade 4.8) may not be identical between the two membranes as discussed in further detail below. Permeance values can vary significantly depending on chemical composition of the membrane, the size of the permeating molecule, the thickness of the membrane—even the membrane manufacturing process or operational conditions (e.g., partial pressures of specific gases exposed to the membrane) can affect the membrane's permeance for one type of molecule versus another. For example, a POSITA would have known that CO₂ can plasticize cellulose acetate membranes and that the permeability (and, thus, permeance) of cellulose acetate is sensitive to the history of how the membrane is exposed to partial pressures of CO₂. EX2014-

*Ex. 2008, Geise Decl., ¶70
POR, 32-33*

A POSITA Would Not Have Looked to Scholz Figure 7



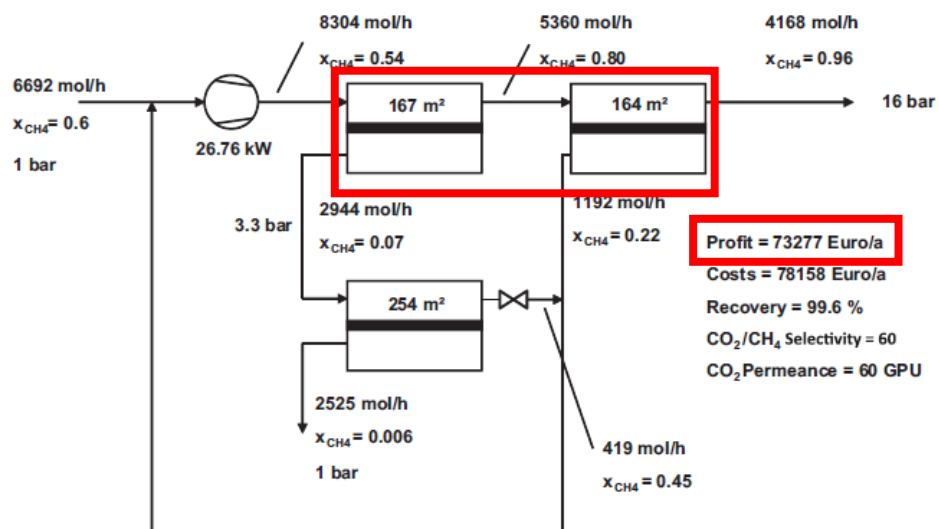
Q. So the methane recovery using Scholz's figure 7A would be lower than the methane recovery just using comparative example one of Ungerank as it already exists, correct?

A. Yes. Yes.

Ex. 2018, Baker May Dep., 45:21-47:24

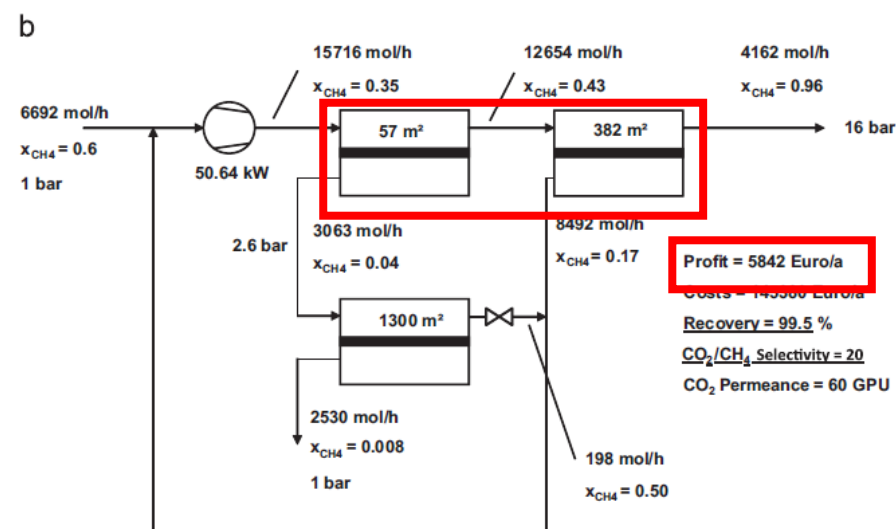
A POSITA Would Not Have Looked to Scholz Figure 7

Scholz Figure 5



Ex. 1007, Figure 5

Scholz Figure 7(b)



Ex. 1007, Figure 7(b)

Petitioner's Motivation to Combine Theory is Legally Insufficient

St. Jude Med., LLC v. Snyders Heart Valve LLC

“Board’s determination that St. Jude did not adequately or persuasively establish the motivation to make the particular combination it proposed in arguing obviousness.”

977 F.3d 1232, 1243 (Fed. Cir. 2020)

Samsung Elecs. Co. v. Dynamics Inc.

No motivation to combine where the proposed combination “would not improve the functionality” of the proposed base reference.

No. 2021-2352, 2022 WL 3041158, at *3 (Fed. Cir. Aug. 2, 2022)

Ungerank Comparative Example 1 is Functionally Complete



Q. But I think what you just said is that somebody looking and reading at Ungerank Comparative Example 1 would understand that that is an operative system that accomplishes its objective, correct?

A. That's correct.

Q. And you — they wouldn't have seen some deficiency in that system, correct?

A. There's nothing there that jumps out at you as a problem.

*Ex. 2009, Baker Dec. Dep., 113:21-114:8;
POR, 42-43*

No Rationale for Selecting Figure 7(a) or 7(b) of Scholz

Figure 5

Profit = 73277 Euro/a
Costs = 78158 Euro/a
Recovery = 99.6 %

Figure 7a

Profit = 61966 Euro/a
Costs = 89485 Euro/a
Recovery = 98.7 %

Figure 8a

Profit = 86238 Euro/a
Costs = 64981 Euro/a
Recovery = 99.5 %

Figure 7b

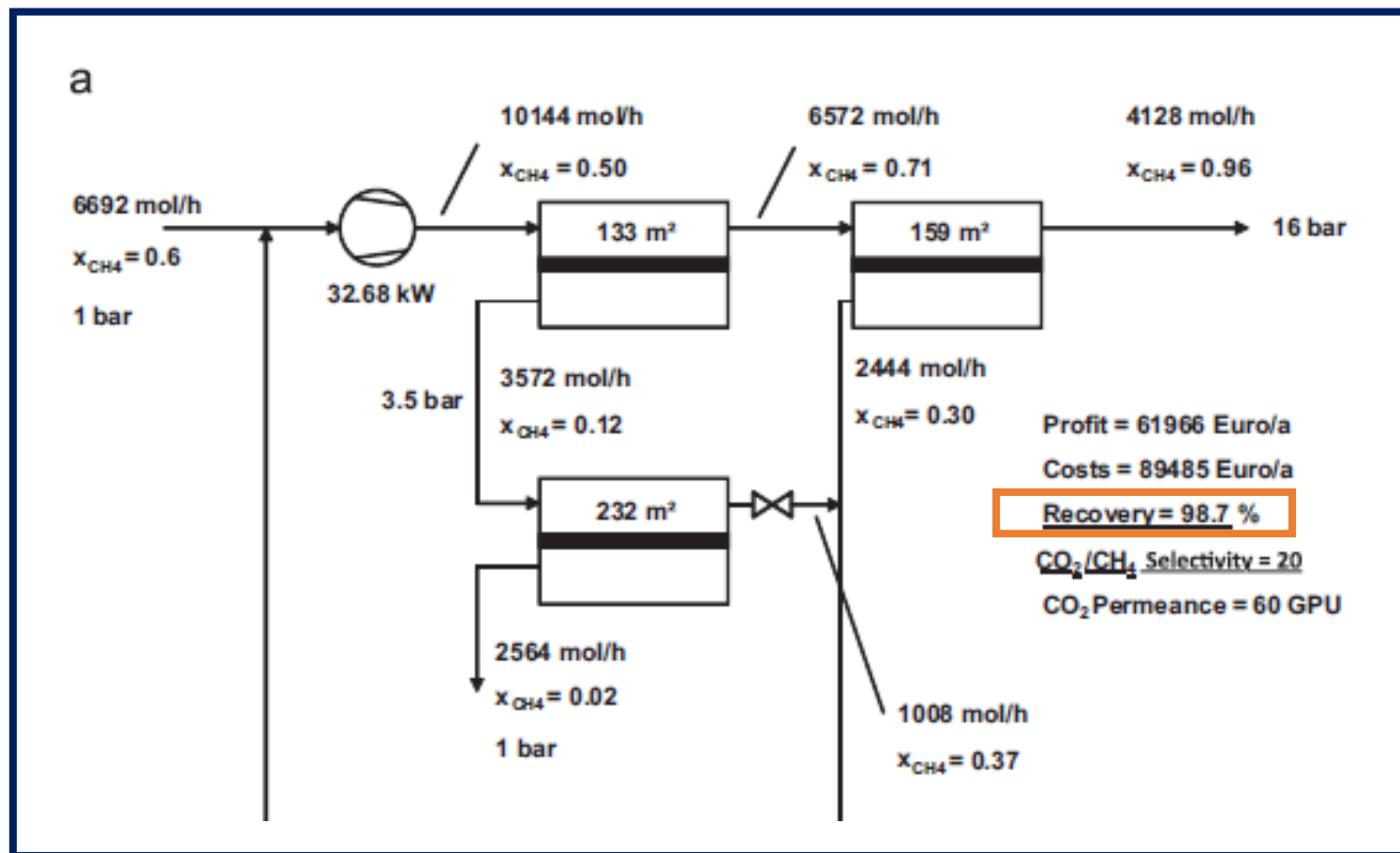
Profit = 5842 Euro/a
Costs = 145380 Euro/a
Recovery = 99.5 %

Figure 8b

Profit = 86007 Euro/a
Costs = 65212 Euro/a
Recovery = 99.5 %

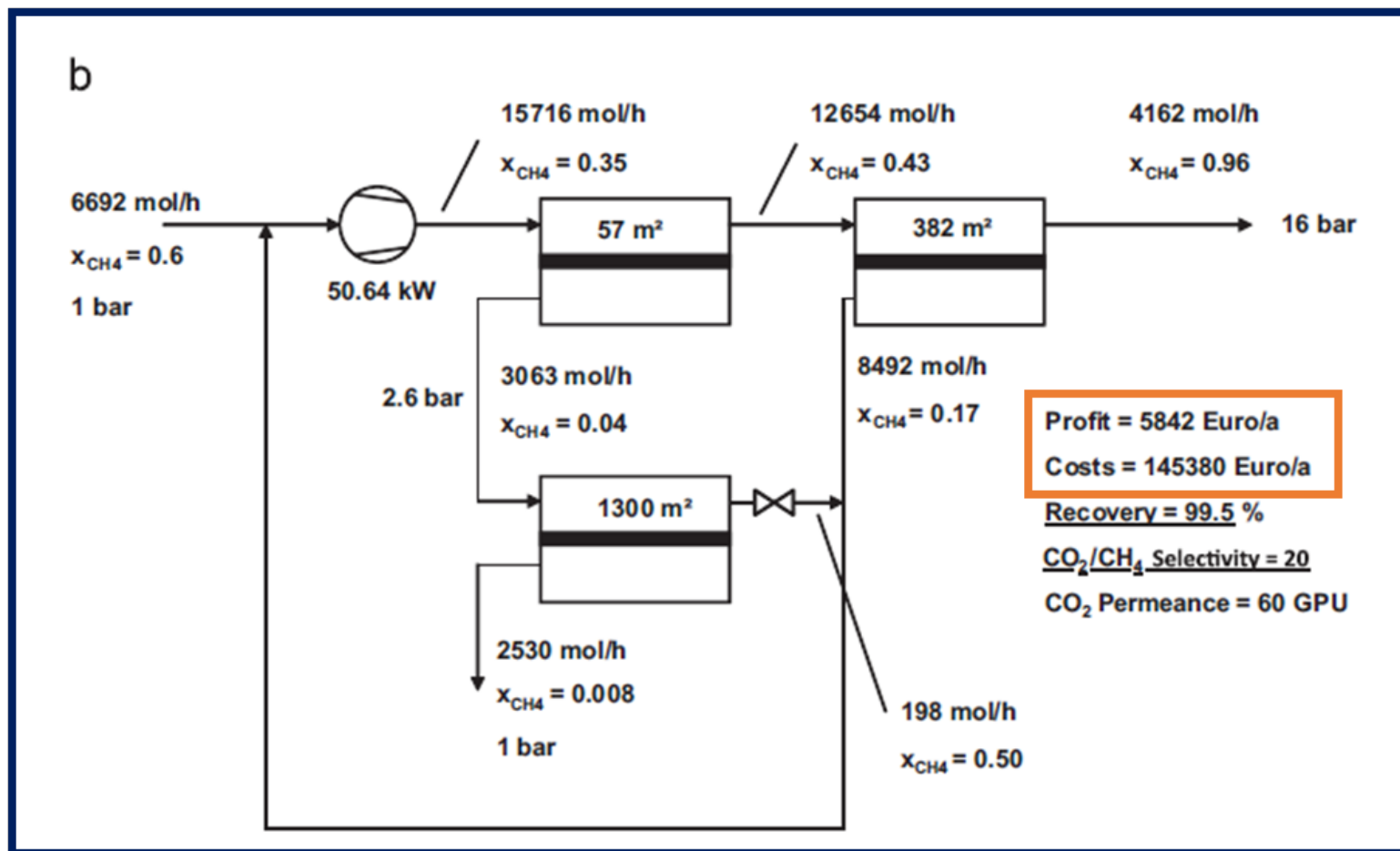
Scholz, FIG. 5, 7(a), 7(b), 8(a), 8(b)
POR, 45

Figure 7(a) of Scholz Has Unacceptable Purity/Recovery Levels



Scholz, FIG. 7(a)
POR, 46-47

Figure 7(b) Has Higher Costs and Lower Profitability



Scholz, FIG. 7(b)
POR, 47

Petitioner's New Obviousness Theory Is Also Incorrect

Petitioner's New Theory Regarding UCE1

The fact that Scholz's Figure 7 systems had larger stage 2 membrane areas than stage 1, and that was consistent with POSITAs' likely intuition about UCE1's stage 2 and stage 1 areas, was an additional reason POSITAs would have found it appropriate to look to Scholz's Figure 7 to better understand UCE1's relative surface areas. AP-1041, ¶¶31–32.

Petitioner's New Theory Regarding UCE1

As Baker explained, POSITAs "might have already figured out" that in UCE1, stage 2's surface was larger than stage 1's area. EX2009, 111:3–113:6. POSITAs would have understood that smaller membranes result in increased permeate concentration, and in UCE1, a high permeate (CO₂) concentration was needed in stage 1 (i.e., in stream (6)) to create the driving force necessary in stage 3 to achieve the relatively high CO₂ concentration (99.0%) in the third permeate stream (11). AP-1041, ¶¶28–30. POSITAs also would have understood that in stage 2, a relatively large membrane was needed to allow much of the CO₂ to pass through, to achieve the relatively high methane concentration (98.5%) in the second retentate stream (8). *Id.*

Reply, 19

The fact that Scholz's Figure 7 systems had larger stage 2 membrane areas than stage 1, and that was consistent with POSITAs' likely intuition about UCE1's stage 2 and stage 1 areas, was an additional reason POSITAs would have found it appropriate to look to Scholz's Figure 7 to better understand UCE1's relative surface areas. AP-1041, ¶¶31–32.

Reply, 19

29. A POSITA would have understood that in Ungerank's Comparative Example 1, to achieve the relatively high concentration of CO₂ (99.0%) in the third permeate stream (11), a relatively large driving force would be needed in stage 3, meaning there must be a relatively high concentration of CO₂ in the first permeate stream (6). A POSITA would therefore have understood that stage 1's membrane must be relatively small.

Ex. 1041, Baker Supp. Decl., ¶ 29

30. For stage 2, the same principle applies. However, the second permeate stream (9) is recycled. In other words, a POSITA would have understood that in Ungerank's Comparative Example 1, to achieve the relatively high concentration of methane (98.5%) in the second retentate stream (8), a relatively large membrane would be needed in stage 2 to allow much of the permeable component (CO₂) to pass through it. A relatively large membrane in stage 2 would also allow a relatively large amount of methane to pass through stage 2's membrane into the second permeate stream (9), but that stream, including the methane therein, is recycled and combined with the crude gas stream (17) so that it processed again.

Ex. 1041, Baker Supp. Decl., ¶ 30

Petitioner's New Theory of Obviousness Is Untimely and Should Be Disregarded

Intelligent Bio-Systems, Inc. v. Illumina Cambridge Ltd.

“The Board did not err in refusing the reply brief as improper under 37 C.F.R. § 42.23(b).”

821 F.3d 1359, 1370 (Fed. Cir. 2016)

Appeal Board Consolidated Trial Practice Guide

“[A] reply or sur-reply that raises a new issue or belatedly presents evidence may not be considered . . . ‘Respond,’ in the context of 37 C.F.R. § 42.23(b), does not mean proceed in a new direction with a new approach.”

Nov. 2019 CTPG, 74

Acceleration Bay, LLC v. Activision Blizzard Inc.

The Board did not err in refusing to consider portions of a reply declaration “rais[ing] a new obviousness argument ... that could have been made in the petition.”

908 F.3d 765, 775 (Fed. Cir. 2018)

Petitioner's New Theory Contradicts the Record



Q. And so you understand that reference example one in the Prieske '380 patent is Ungerank's comparative example one, correct?

A. That's my belief, yes.

Ex. 2018, Baker May Dep., 29:21-25

Reference Example 1

The Comparative-Example of WO 2012/000727 A1 was reproduced. Characteristics of this example are:

A recycled gas volume in sum of 86.7%

Identical membrane capacities in all three separation stages

A quotient of the pressure ratio over the permeate separation stage (3) to the pressure ratio over the feed stream separation stage (1) of 1.0.

Membrane selectivity for $\text{CO}_2/\text{CH}_4=20$ in all separation stages

Thus, the reference example differs from the present invention in that the membrane capacities are identical in all three stages.

Ex. 1001, '380 Patent, 16:21-25

Sur-Reply, 8-9

Petitioner's New Theory Contradicts the Record



Q. So that's contrary to the intuition you had in reading Ungerank compared to example one, correct?

MR. LANE: Objection.

Form.

A. Yes. I mean, I'm not saying that — that it's possible to make a process where all three stages are equal and it will converge and it will — it may give you a fairly pure CO₂ and fairly pure methane.

Ex. 2018, Baker May Dep., 31:5-14

A POSITA Would Not Confirm an “Intuition” using Scholz



Q. Now, one difference between the membranes used in comparative example one of Ungerank and Scholz figure 7A is that the Ungerank membranes are polyamide fibers and the figure 7A of Scholz membranes are cellulose acetate, correct?

MR. LANE: Objection.

Form.

A. That's correct.

* * * * *

So the relative areas of the polyamide and the Scholz membrane — of the CA membrane — may be different because their permeances may be different.

*Ex. 2018, Baker May Dep., 50:5-51:7
Sur-Reply, 10-11*

Petitioner Mischaracterizes the Problem to Be Solved

Petitioner Mischaracterized the Problem Solved by Priske-'380

Energy and membrane costs are two of the most significant costs in building and operating membrane-separation systems, and a POSITA would have understood before the Critical Date there are tradeoffs between the two. *See, e.g.*, AP-1013, 346; AP-1003, ¶¶120-127. Ungerank explained that “[m]embranes of

* * *

In other words, a POSITA would have understood the cost tradeoffs between lower-selectivity and higher-selectivity membranes: higher-selectivity membranes are more expensive than low-selectivity membranes, but higher-selectivity membranes result in lower recycle volumes, requiring less compression, which requires less energy, which can be costly. AP-1003, ¶¶122-123.

Purported Trade-offs Would Not Lead to Claimed Invention



72. Dr. Baker generally alludes to “trade-offs” between “energy and membrane replacement costs” as the problem motivating a person of ordinary skill to arrive at the claimed invention. AP1003-Baker Decl., ¶81. This is not the problem identified in the Priske-’380 patent and even if it was, it would not lead a person of ordinary skill to the claimed invention.

*Ex. 2008, Geise Decl., ¶72
POR, 34*

Priske-'380 is Directed to a New Problem in Gas Separation

In recent times, however, a new problem has occurred, that is not solved in a sufficient manner by the apparatuses and processes of the prior art. The problem is that some gas separation plants are operated at sites where energy is very cheap. At such locations the focus of operators of separation plants lies on investment and maintenance costs rather than on an energy saving. No sufficient solution for this problem has been found in the prior art.

*Ex. 1001, '380 Patent, 2:27-30
POR, 34-35*

Petitioner Has Not Established That This Problem Was Recognized in the Prior Art

Purported Evidence

125. For the reasons explained above, it would have been obvious to a POSITA in December 2014 to use lower-selectivity, higher permeance (and therefore smaller area and less costly) membranes in that situation. For example, I described in my book *Membrane Technology and Applications* how membrane systems had been installed on offshore platforms, where small footprint and low weight are important. AP-1013, 363. On offshore platforms, low-cost energy is usually available, for example, hydrocarbon gases that otherwise might be flared.

Baker's Testimony

Q. And — and there's no reference in — in this portion of your textbook to the selectivity or the permeance of the membranes that are used, correct?

A. Not in this portion, but elsewhere there probably is. But at least not — I don't see it in this portion. A

Q. And this section of your textbook doesn't say anything about the cost of energy, correct?

A. No. Not that I can recall.

Ex. 1003, Baker Decl., ¶125
Ex. 2009, Baker Dec. Dep., 164:4-7
POR, 34-35

Where Energy Cost is Different, Not Addressed in the Art



75. Further, the Ungerank patent and Scholz do not address scenarios where the energy cost is different. Scholz assumes a fixed cost of 0.08 euros per year, and Ungerank is silent as to electrical cost. *See* AP1007-Scholz, Table 3.

76. Scholz states that “operational cost” is “mainly caused by electrical energy consumption.” AP1007-Scholz, 4. This statement indicates that Scholz

77. The Ungerank patent does not discuss scenarios where lower energy cost is present. Instead, the Ungerank patent focuses on the benefits of high selectivity membranes because these allow the separation to “be conducted in a more economically viable manner,” because “the necessary size of the compressor and the energy required can be reduced.” AP1006-Ungerank patent, 9:30-34.

Scholz Does Not Contemplate Low Energy Costs

Table 3

Economic parameters used for optimization of a membrane based biogas upgrading plant.

Costs	Unit	Value
Membrane module costs	$\frac{\text{Euro}}{\text{m}^2}$	55
Electrical costs	$\frac{\text{Euro}}{\text{kWh}}$	0.08
Commission of purified gas	$\frac{\text{Euro}}{\text{m}^3}$	0.2
Membrane lifetime	year	4
Annual operation	h	8,000
Interest rate	–	0.09
Amortization period	years	8

Ex. 1007, Scholz, 4
POR, 36

Without a Recognized Problem the Petition Fails

Forest Laboratories, LLC v. Sigmapharm Laboratories, LLC

“[W]here a problem was not known in the art, the solution to that problem may not be obvious, because ordinary artisans would not have thought to try at all because they would not have recognized the problem.”

817 F.3d 928 (Fed. Cir. 2019)

Virteck Vision Int’l ULC v. Assembly Guidance Systems, Inc.

Substantial evidence did not support finding that prior art references provided motivation to combine where there was “no evidence of a design need or market pressure.”

97 F.4th 882 (Fed. Cir. 2024)

**Petitioner Has Provided No Rationale For Why A POSITA
Would Start With Ungerank CE1**

Ungerank Teaches that CE1 is Not Economically Favorable

COMPARATIVE EXAMPLE 1

Separation of a Mixture of Methane Dioxide with a Mixing Ratio of 50 Moderately Selective Polyimide

The FIG. 11 shown connection arrangement was used. Each stage consisted of a hollow fiber membrane module consisting of hollow polyimide fibers from UBE (NM B01 A type). 1.78 m³/h of a crude gas mixture of 50% methane and 50% carbon dioxide, which corresponds approximately to a gas mixture of biogas, is introduced into a mixing chamber (not shown in FIG. 11) and then compressed to 25 bar together with recycled gas composed of gas streams (9) and (10). The gas which has been compressed and cooled to 20° C. is applied to the feed stream separation stage (1). The retentate of the first separation stage is then passed to the second separation stage (2) on the other side of the retentate stream of the first stage, thus determining the methane content of the retentate stream of the second membrane separation stage (2) to be 1.5% carbon dioxide. The retentate stream of the second separation stage (2) is then passed to the third separation stage (3) on the other side of the retentate stream of the second stage, thus determining the methane content of the retentate stream of the third separation stage (3) to be 65.5%, and is recycled via the second permeate stream (7) into the mixing chamber and compressed again by the compressor (4).

11
EXAMPLE 1

compression rate. The membranes used exhibit a moderate mixed gas selectivity for carbon dioxide over methane of 20.

The FIG. 11 shown connection arrangement was used. Each stage consisted of a hollow fiber membrane module consisting of highly selective hollow polyimide fibers with a separation area of about 5 m² per module. These hollow polyimide fibers were produced according to example 19 of Austrian patent application A1164/2009, except working with a precipitation bath temperature of 40° C. rather than 10° C. 1 m³/h of a crude gas mixture of 50% methane and 50%

rate. The membranes used exhibit a high mixed gas selectivity for carbon dioxide over methane of 45. The high selectivity of 45 compared to 20 in the comparative example therefore enables a more economically favorable process by a lower recompression of 31.2% rather than 86.7%.

bonds approximately to a gas ed into a mixing chamber and zether with recycled gas com- (10). The gas which has been C. is applied to the feed stream ate of this stage is passed via the retentate separation stage e retentate side of the retentate 4 bara and thus determines the brane of membrane separation ate of the retentate separation

Ex. 1006, Ungerank Patent, CE1, Example 1 (compared)
POR, 39-40

CE1 Results in an Undesirable Amount of Methane Loss

COMPARATIVE EXAMPLE 1

Separation of a Mixture of Methane and Carbon Dioxide with a Mixing Ratio of 50 to 50 by Moderately Selective Polyimide Membranes

The FIG. 11 shown connection arrangement of the separation stages (1) and (2) is shown in FIG. 11. Each stage consisted of a hollow fiber membrane module consisting of hollow polyimide fibers from UBE (NM B01 A type). 1.78 m³/h of a crude gas mixture of 50% methane and 50% carbon dioxide, which corresponds approximately to a gas mixture of biogas, is introduced into a mixing chamber (not shown in FIG. 11) and then compressed to 25 bar together with recycled gas composed of gas streams (9) and (10). The gas which has been compressed and cooled to 20° C. is applied to the feed stream separation stage (1). The retentate of the feed stream separation stage (1) is subsequently passed via the first retentate stream (7) to the separation stage (2). A reducing valve (13) on the side of the retentate separation stage (2) is set to 18.2 bar, thus determines the driving force through the membrane separation stages (1) and (2). The retentate of the separation stage (2) has a content of 98.5% methane and 1.5% carbon dioxide. 0.895 m³/h of this mixture is applied to the separation stage (2). The permeate of the separation stage (2) has a volume flow rate of 0.743 m³/h, a methane content of 34.5% and a carbon dioxide content of 65.5%, and is recycled via the second permeate stream (10) into the mixing chamber and compressed again by the compressor (4).

separation stage (3). This gives a third permeate stream (11) from permeate separation stage (3) of 0.885 m³/h with a composition of 99.0% carbon dioxide and only 1.0% methane. The third retentate stream (10) from the permeate separation stage (3) is recycled via the third retentate stream (10) into the mixing chamber and compressed again by the compressor (4).

Ex. 1006, Ungerank Patent, 10:57-59

In order to satisfy various legislators, it is necessary to minimize the methane loss into the atmosphere. The FIG. 11 shown connection arrangement was used to lower the concentration of methane in the third permeate stream (11) to below 0.5% by volume. Each stage consisted of a hollow fiber membrane module consisting of hollow polyimide fibers from UBE (NM B01 A type).

Ex. 1006, Ungerank Patent, 11:62-63

Ungerank CE1 is not a Commercially Useful Starting Point



significant figures, and the flow rates are very small — well, less than a meter cubed per hour.

That's a pretty — that's a — that's almost a lab scale system. I'm not

...

Well, it's treating — the total amount of gas it's treating is less than 2 cubic meters per hour. That's a pretty small amount of gas.

...

A. A commercial system is treating a few hundred, I mean, cubic meters per hour. Tens to hundreds, several you can see.

Q. But it is a — it is a very significantly larger system than what we're seeing in Comparative Example 1, correct?

A. Absolutely.

MR. LANE: Objection to form.

A. Yes. Yes. Example 1 was small, tiny.

*Ex. 2009, Baker Dec. Dep., 95:8-97:24, 126:19-127:11
POR, 40*

Ungerank CE1 is not a Commercially Useful Starting Point



23. A POSITA would have understood that with the crude gas flowrate of 1.78 m³/hr in Ungerank's Comparative Example 1, any type of commercial membrane modules would have been far too large for that system.

*Ex. 1041, Baker Supp. Decl., § 23
POR, 40*

Membranes Used in Ungerank CE1 Were Uncommon



Q. And the material used in Comparative Example 1 is relatively uncommon in the market, correct?

MR. LANE: Objection. Form.

Objection. Misstates his testimony.

A. It's -- it has a fraction of the market, but not a big fraction. Just to clarify that.

Petitioner's Rationale for Starting With UCE1 Could Just as Well Apply to Scholz



82. Dr. Baker asserts four reasons that a person of ordinary skill, in his view, would start with Ungerank CE1 when designing a new process: (1) it has low selectivity membranes, (2) it produces relatively pure product streams, (3) it is a three stage system with a single compressor, and (4) it provides detailed information on the process parameters including gas composition, flowrates, recycle volume, pressures, and it specifies the types of membrane used. AP1003-Baker Decl., ¶¶131-133; EX2009-Baker Dep., 105:18-107:4. However, Dr. Baker admitted that the systems in Scholz Figure 7 have all of these characteristics as well. EX2009-Baker Dep., 108:3-24. As a result, Dr. Baker has not identified any specific reason that a person of ordinary skill would have started with CE1 when designing a new separation process.

No Reasonable Expectation of Success

Petitioner's Argument is Legally Defective

then be optimized. *Id.*, ¶138. The detailed system information in Ungerank and Scholz would have provided a POSITA confidence that a design based on these teachings would work. *Id.*

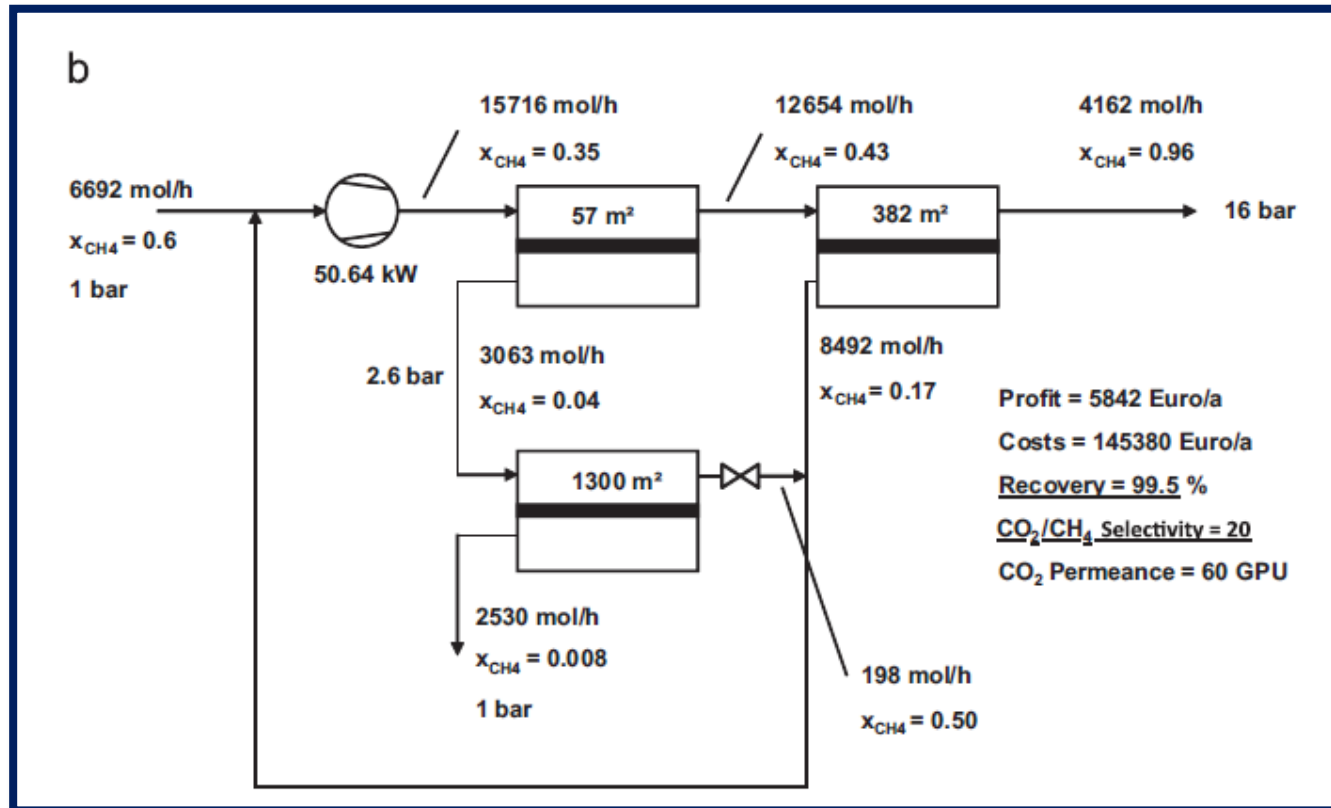
Corrected Petition, 24

Eli Lilly & Co. v. Teva Pharms. Int'l GmbH

“We ... emphasize the clear distinction in our case law between a patent challenger’s burden to prove that a skilled artisan would have been motivated to combine prior art references and ***the additional requirement that the patent challenger also prove that the skilled artisan would have had a reasonable expectation of successfully achieving the claimed invention from the combination.***”

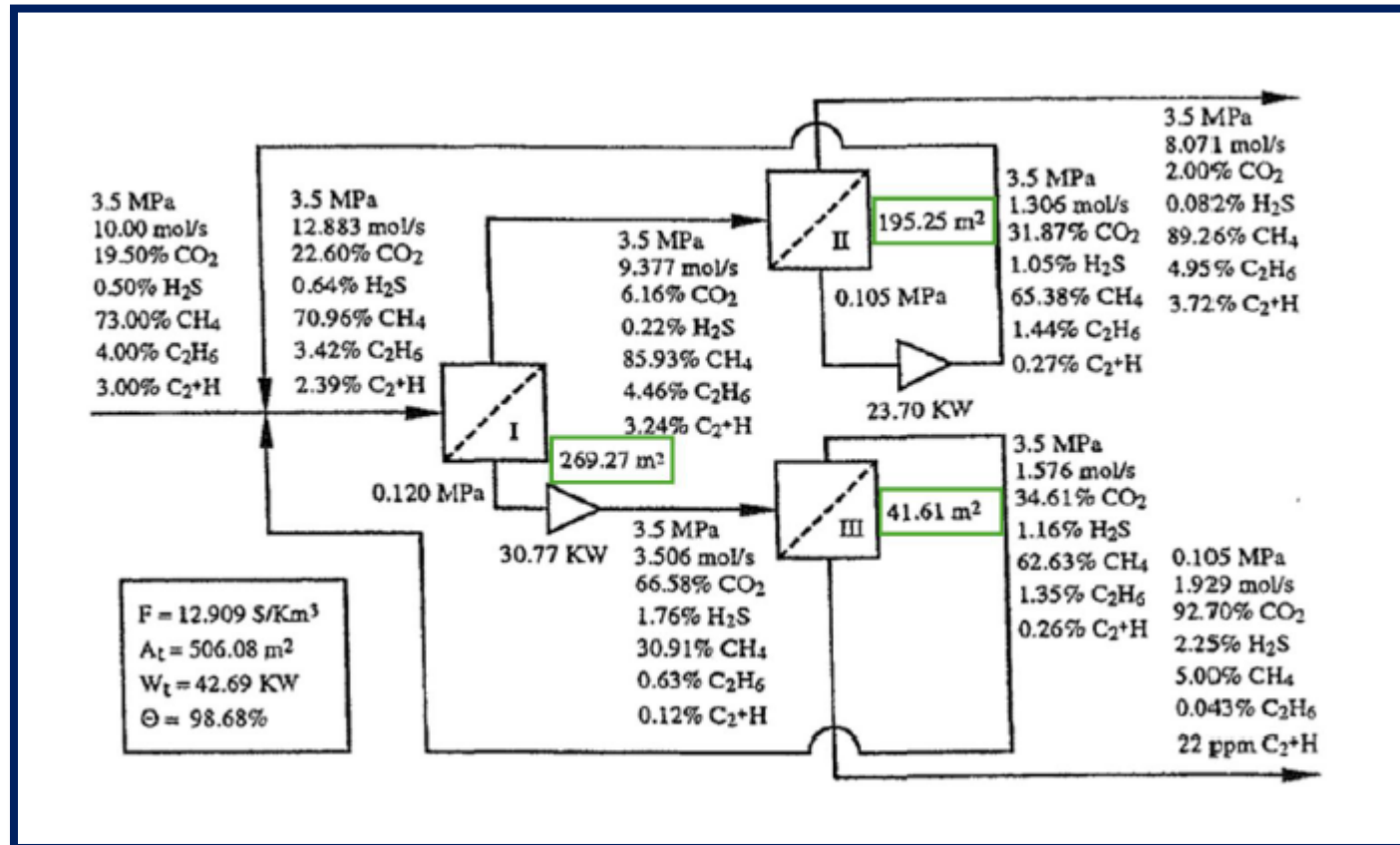
8 F.4th 1331, 1344 (Fed. Cir. 2021)

Optimization of Membrane Separation Systems Depends on a Large Number of Interdependent Variables



Ex. 2007, Scholz, FIG. 7(b)
POR, 48-49

Optimization of Membrane Separation Systems Depends on a Large Number of Interdependent Variables



Ex. 1009, Qi, FIG. 9
POR, 48-49

Optimization of Membrane Separation Systems Depends on a Large Number of Interdependent Variables



97. Given the large number of variables that could be modified, and that the effect of each of these variables on system optimization may depend in a complex manner on a number of other variables, assumptions or other design constraints—absent further guidance—a person of ordinary skill in the art would not have had a reasonable expectation of success at selecting the surface membrane ratios taught in Figures 7(a) and/or 7(b) for combination with another system with a reasonable expectation of success.

*Ex. 2008, Geise Decl., ¶197
POR, 48-49*

No Reasonable Expectation of Success

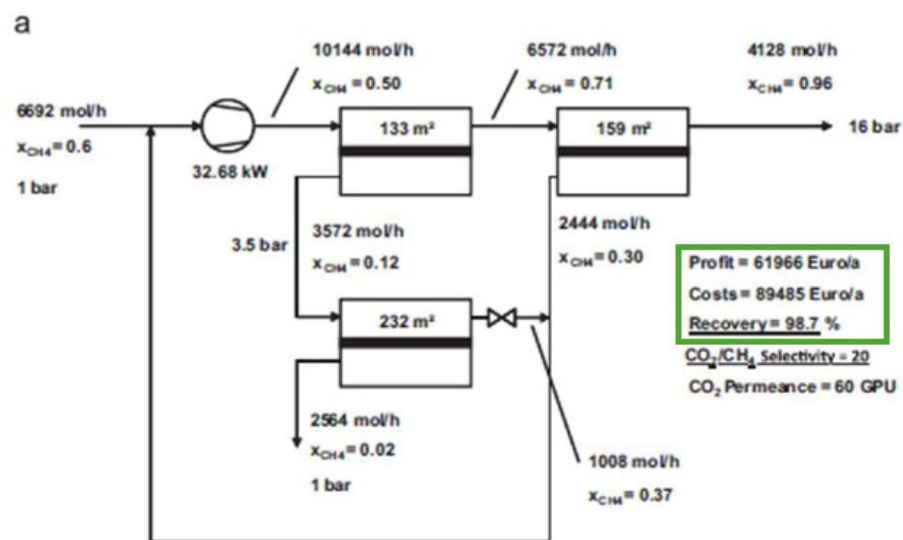
Pfizer, Inc. v. Apotex, Inc.,

“To have a reasonable expectation of success, one must be motivated to do more than merely to vary all parameters or try each of numerous possible choices until one possibly arrived at a successful result.”

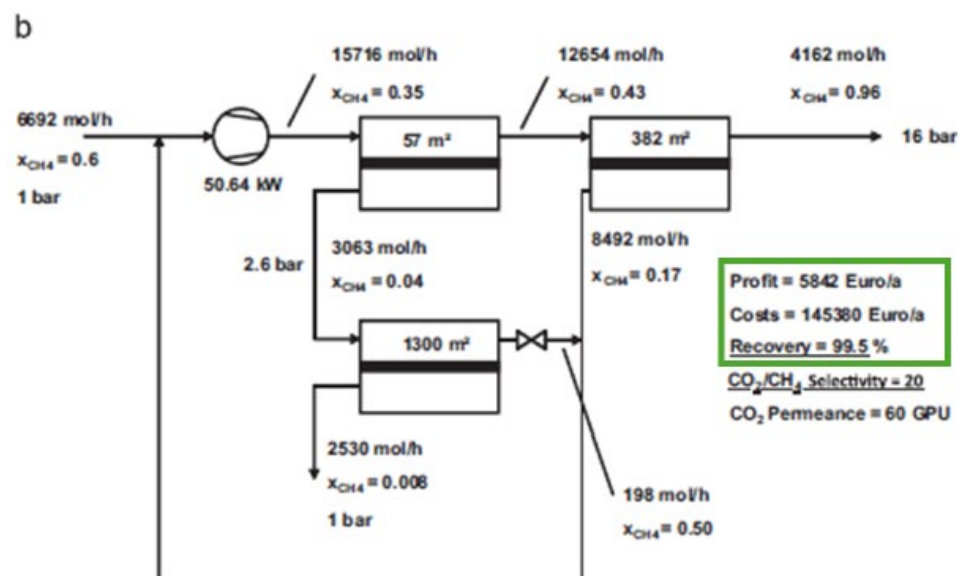
480 F.3d 1348, 1365 (Fed. Cir. 2007)

Even Small Changes Can Lead to Large Differences in Profits, Costs and Percent Recovery

Scholz Figure 7(a)



Scholz Figure 7(b)



Scholz Model Assumes Ideal Gases, Not Real World Applications



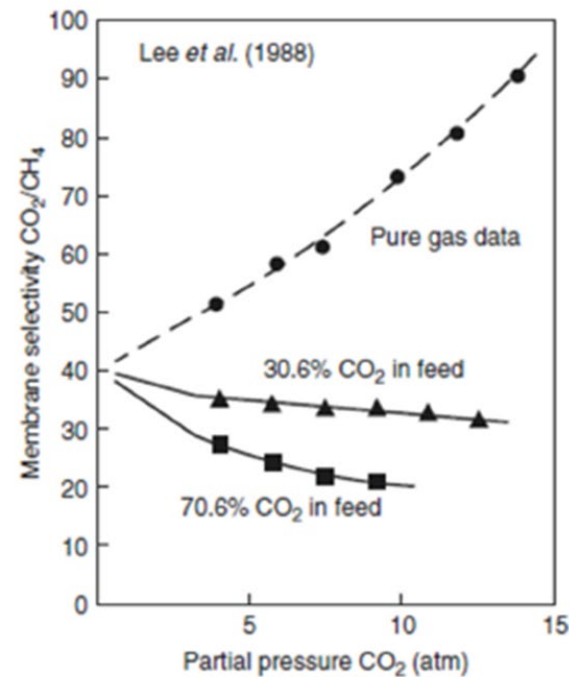
101. Moreover, the Scholz model assumes that the system involves ideal gases—a condition rarely encountered in real-world applications involving CO₂ and CH₄—and does not account for concentration polarization or variations in selectivity for mixed gas transport, both of which are critical factors in the separation of mixed gas systems. Gas concentration polarization is the phenomena “[w]hen a mixture is brought to a membrane surface by any driving force, there is an accumulation of the less permeable species and a depletion of the more permeable components in the boundary layer adjacent to the membrane and this causes a concentration gradient building up in the boundary layer.” EX2015-He, 243. Concentration polarization can lead to performance losses by creating concentration gradients that reduce the driving force for separation, and changes in membrane selectivity can significantly impact the efficiency and effectiveness of the membrane in a gas separation process.

*Ex. 2008, Geise Decl., ¶101
POR, 52-53*

Mixed Gases and Pure Gases Can Have Large Differences in Selectivities



102. The literature suggests that the difference between selectivities calculated from pure gas measurements and *selectivity values measured using mixed gas measurements* can be large. AP1013-Baker Textbook, 345 (citing Lee,



Ex. 2008, Geise Decl., ¶102
POR 52-53

No Reasonable Expectation of Success



As can be seen from the Lee et al. study, the selectivity measured with a CO₂/CH₄ mixture can be one-half or less of the selectivity calculated from pure gas measurements. A person of ordinary skill in the art, would understand that failing to account for these differences results in an overestimation of the ability of a membrane to separate a target gas mixture. Thus, a person of skill in the art would not have simply expected the calculated membrane areas and the given selectivities to have successfully translated to a system that was practically combinable with Ungerank with any expectation of success.

*Ex. 2008, Geise Decl., ¶102
POR 52-53*

No Reasonable Expectation of Success

UCE1 does not provide the membrane surface areas, leaving POSITAs a finite number of identified options in terms of stage 2's relative size versus stage 1. *KSR*, 550 U.S. at 421. Stage 2 could be larger, equal to, or smaller than stage 1. As explained above, Scholz's Figure 7 (as well as UCE1 itself) would have led to the belief that stage 2 was larger than stage 1.

Given the finite options, POSITAs would have had a reasonable expectation of success using Scholz's Figure 7, in conjunction with the information in UCE1 itself, to understand that in UCE1, stage's 2's surface area was larger than in stage 1. Using Scholz's Figure 7 in this way would not have involved changing any

Claims 9 and 12 are Not Obvious

Dependent Claim 9 Is Not Obvious

9. The method of claim 1, wherein the combined gas volume recycled with the second permeate stream and the third retentate stream is from 65 to 85% of the volume of the crude gas stream.

Ex. 1001, Priske-'380, claim 9

mixing chamber and compressed again. The sum of recycled gas streams (9) and (10) is therefore 1.544 m³/h or 86.7% based on the amount of gas to be separated supplied. Pure

Ex. 1006, Ungerank, 10:62-64

In re Patel

“[P]roximity alone is not enough to establish a prima facie case of obviousness.”

566 F. App'x 1005, 1009 (Fed. Cir. 2014)

Sur-Reply, 21-22

Dependent Claim 12 is Not Obvious

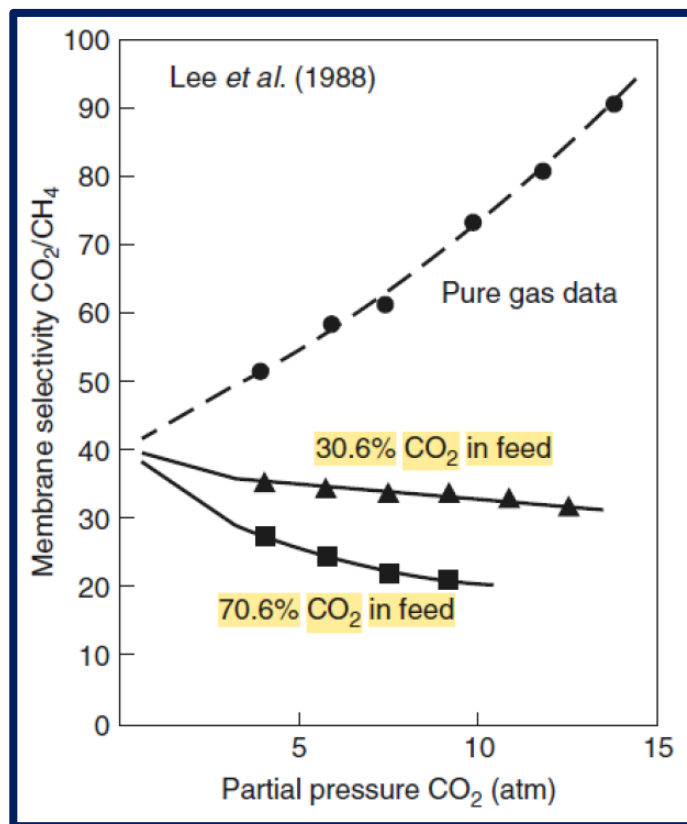
12. The method of claim 11, wherein the gas separation membranes in the feed stream separation stage have a pure gas selectivity for CO₂/CH₄ of from 20 to 100.

Ex. 1001, Priske-'380, claim 12

compression rate. The membranes used exhibit a moderate mixed gas selectivity for carbon dioxide over methane of 20.

Ex. 1006, Ungerank Patent, 10:66-67

Dependent Claim 12 is Not Obvious



Ex. 1013, Baker Textbook, Figure 8.8 (annotated)

- Data pertains to cellulose acetate membranes, not polyimide membranes as in UCE1.

Ex. 1013 (Baker Textbook), 345, Figure 8.8

Ex. 1006, Ungerank, 10:66-67

- No data for 61.2% CO_2 stream as in UCE1.

Ex. 1006, Ungerank Patent, 10:15-67

Dependent Claim 12 is Not Obvious

citing EX2008, ¶¶108–109. Regardless, in 2010 Bhatti reported CO₂/CH₄ pure-gas selectivities for polyimide membranes of 29 to 59—nowhere near five times the mixed-gas selectivity of 20 in UCE1’s polyimide membranes, and in 1999, Staudt-Bickel reported similar data for polyimide membranes. AP-1048, 5; AP-1049, 6, 8; AP-1041, ¶¶50–55.

Reply, 26

Appeal Board Consolidated Trial Practice Guide

“[A] reply or sur-reply that raises a new issue or belatedly presents evidence may not be considered.”

Nov. 2019 CTPG, 74

Sur-Reply, 22-23

Ground II

Petitioner Does Not Address Ground II in Its Reply

TABLE OF CONTENTS

I.	INTRODUCTION	1
II.	THE PARTIES' EXPERTS	2
	A. Evonik Has No Expert That Has A POSITA's Qualifications	2
	B. Petitioner's Expert Has A POSITA's Qualifications.....	3
III.	PRIOR ART	4
IV.	CLAIM CONSTRUCTION: SCHOLZ'S AND UCE1'S CAPACITY RATIOS AND SURFACE AREA RATIOS ARE EQUAL	6
V.	UNGERANK IS PRIOR ART	9
VI.	POSITAS WOULD HAVE BEEN MOTIVATED TO USE LOWER- SELECTIVITY MEMBRANES	11
VII.	EVONIK'S ARGUMENTS THAT PRIOR ART HAD TO ADDRESS THE "NEW PROBLEM" IS WRONG.....	13
VIII.	POSITAS WOULD HAVE LOOKED TO UCE1	15
IX.	POSITAS WOULD HAVE BEEN MOTIVATED TO COMBINE UCE1'S AND SCHOLZ FIGURE 7'S TEACHINGS, THEREBY MEETING CLAIM 1's CAPACITY LIMITATION.....	18
X.	POSITAS WOULD HAVE HAD A REASONABLE EXPECTATION OF SUCCESS.....	23
XI.	DEPENDENT CLAIMS	25

Petitioner Does Not Address Ground II in Its Reply



Q. Now, in your supplemental declaration, you don't offer any opinion that the Scholz article alone renders the claims of the Priske patent obvious, correct?

A. I don't do that. That's true.

Ex. 2018, Baker May Dep., 12:24-13:5

Scholz Does Not Render Obvious Limitation 1.f.i

i) the combined gas volume of the second permeate stream and the third retentate stream is from 60 to 100% of the volume of the crude gas stream,

Ex. 1001, '380 Patent, claim 1

In Scholz's Figure 7 (a) system “the combined gas volume of the second permeate stream and the third retentate stream” as a percentage “of the volume of the crude gas stream” (i.e. “recycle percentage” or “recycle volume”) is 51%. AP-1007, 7. In the Figure 7(b) system, the recycle percentage is 135%. *Id.*; AP-1003, ¶205. The claimed “60 to 100%” range falls within the two single recycle percentages (51% and 135%) disclosed in Scholz, and thus would have been *prima facie* obvious. AP-1003, ¶¶205-210.

Corrected Petition, 47

Scholz Does Not Render Obvious Limitation 1.f.i

Titanium Metals Corp of Am. v. Banner

Claim: 0.3% molybdenum; 0.8% nickel

Prior art: 0.25% molybdenum; 0.75% nickel

Prior art: 0.31% molybdenum; 0.94% nickel

Prior art alloys had “proportions [that] are so close that prima facie one skilled in the art would have expected them to have the same properties.”

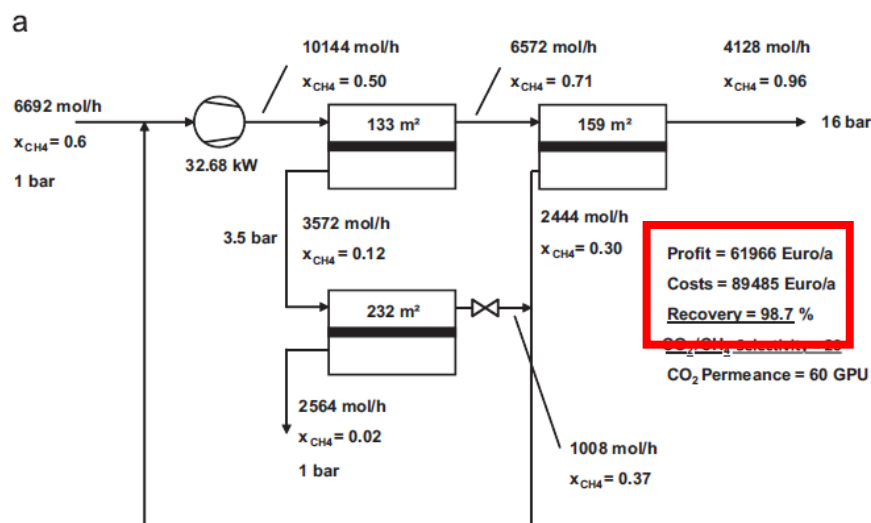
778 F.2d, 783 (Fed. Cir. 1985)

The Panel has some concerns with Petitioner’s analysis regarding the recycling limitation and the pressure ratio limitation. Petitioner, relying on *Titanium Metals*, argues that the “range” of recycling values, Pet. 48, and quotients of the pressure ratios, *id.* at 51–52, taught by Scholz’s Figures 7(a) and (b) are sufficient to support a prima facie case of obviousness.

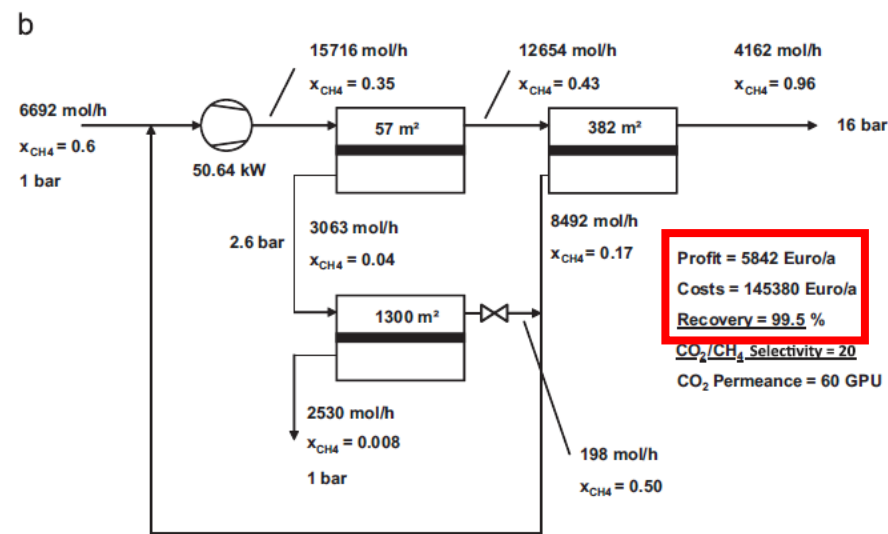
Decision Granting Institution, 27

Scholz Does Not Render Obvious Limitation 1.f.i

Scholz Figure 7(a)



Scholz Figure 7(b)



Ex. 1007, Scholz, Figures 7(a), 7(b)

Scholz Does Not Render Obvious Limitation 1.f.i



Q. -- to 76, in those paragraphs, you don't identify any specific motivation for a person of skill in the art to design a system that achieves a recycle rate specifically of 60 percent or something between 60 and 100 percent as opposed to, for example, 59 percent or 101 percent, correct?

MR. LANE: Objection. Form.

Objection --

A. Yes.

Q. And the person who's testing this system would, for example, also be testing, in your opinion, a recycle percentage of, for example, 115 percent, correct?

MR. LANE: Objection. Form.

A. 135, yeah. I mean, we're into the claims, which is sort of a legal thing. I mean, I'm -- so in the claims, I -- if I -- I can -- if the POSITA is -- is told, can you make a system that's between -- between 80 and 100 percent given this information, he can do it.

If somebody tells him, I need it -- it has to be, for whatever reason, I need 60 to 100, he can do it. He's got all the info he needs.

Scholz Does Not Render Obvious Limitation 1.f.i



118. Third, Dr. Baker has not identified any reasons that a person of ordinary skill in the art would modify the systems of Scholz Figure 7 to achieve a recycle volume in the claimed range. To start, Scholz explains that these are the optimized systems. The models in Scholz Figure 7 have different recovery requirements, 7(a) targets 95% and 7(b) targets 99.5%. Put differently, these recovery targets are the variable that changes between 7(a) and 7(b). Dr. Baker has not suggested that a person of ordinary skill in the art would be motivated to pursue a different methane recovery target. Table 3 of Scholz shows that the sale of methane is the only source of revenue for the process, so lower methane recovery will lead to lower amounts of methane for sale, and thus lower revenue.

Ex. 2008, Geise Decl., ¶ 118

Scholz Does Not Disclose Limitation 1.f.ii

See slides 54-60.

Claim 8 | Scholz Does Not Render Obvious the Claimed Range

8. The method of claim 7, wherein the ratio of the total capacity of the gas separation membranes in the permeate separation stage to the total capacity of the gas separation membranes in the feed stream separation stage is in a range of from 0.8 to 1.

Ex. 1001, Priske-'380, claim 8

Claim 8 – The method of claim 7, wherein the ratio of the total capacity of the gas separation membranes in the permeate separation stage to the total capacity of the gas separation membranes in the feed stream separation stage is in a range of from 0.8 to 1.

See [7], *supra*, explaining Scholz's Figure 7 discloses a range for the 3/1 capacity ratio of 1.7-22.8. That range does not overlap the 0.8-1 claimed range, but the low-end of the Scholz range is near the high-end of the claimed range. AP-1003, ¶231.

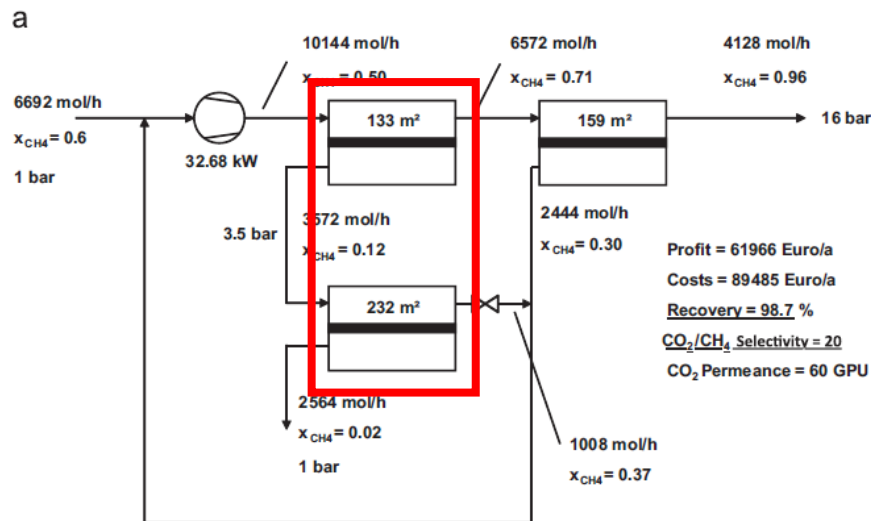
Corrected Petition, 56

Claim 8 | Scholz Does Not Render Obvious the Claimed Range

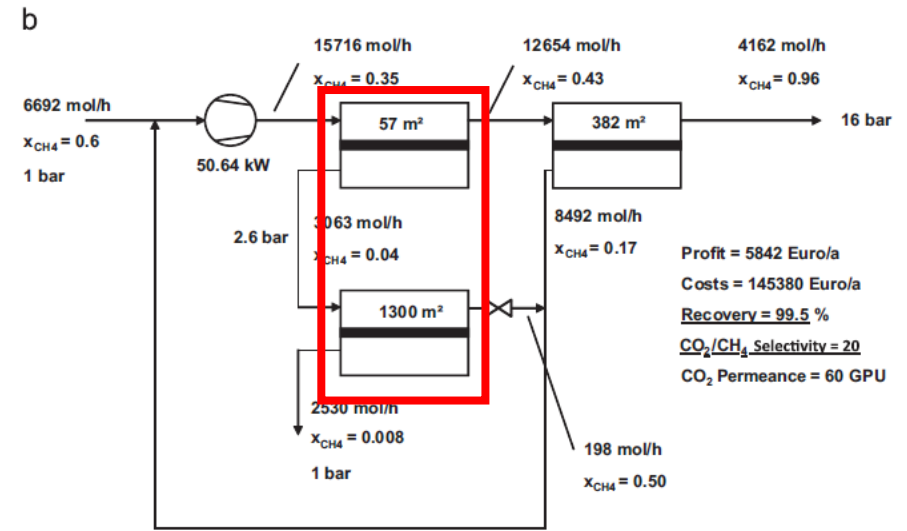
See slides 55-60.

Claim 8 | Scholz Does Not Disclose a “Range”

Scholz Figure 7(a)



Scholz Figure 7(b)



Ex. 1007, Scholz, Figures 7(a), 7(b)

Claim 8 | Scholz Does Not Render Obvious the Claimed Range



125. As Dr. Baker notes, assuming his analysis is correct, the relevant capacity ratio in Scholz 7(a) is 1.7. AP1003-Baker Decl., ¶231. The relevant capacity ratio in 7(b) would be 22.8 ($130/57=22.8$). Dr. Baker apparently suggests that a person of ordinary skill would arrive at the claimed capacity ratio range through mere experimentation. Going from Scholz's closest value to the closest point in the claimed range constitutes a 41% reduction in the capacity ratio ($[1.7-1]/1.7$). While a person of ordinary skill can simulate processes using programs such as Aspen, ChemCad, or GAMS, changing capacity ratios by 41% is more than mere experimentation, it is more like reworking the entire system. Using Scholz as a guide, a person of ordinary skill would understand that the model in 7(a) achieved its goal of 95% methane recovery, and thus would not have a motivation to rework the entire process.

Ex. 2008, Geise Decl., ¶ 125

Claim 9 | Scholz Does Not Render Obvious the Claimed Range

9. The method of claim 1, wherein the combined gas volume recycled with the second permeate stream and the third retentate stream is from 65 to 85% of the volume of the crude gas stream.

Ex. 1001, Priske-'380, claim 9

Claim 9 – The method of claim 1, wherein the combined gas volume recycled with the second permeate stream and the third retentate stream is from 65 to 85% of the volume of the crude gas stream.

See [1.f.i], *supra*. Scholz's Figure 7 discloses a range of 51-135% for the recycle percentage, which encompasses the 65-85% claimed range. AP-1003, ¶232. For the reasons explained for [1.f.i], a POSITA would have found that the recycle percentage range in Scholz's Figure 7 teaches and/or suggests the 65-85% claimed range or at least renders it obvious. *Id.*, ¶¶232-235.

Corrected Petition, 57

Patent Owner's Sur Reply, 21-22

Claim 9 | Scholz Does Not Render Obvious the Claimed Range

See slides 109-113.

Claim 10 | Scholz Does Not Render Obvious the Claimed Range

10. The method of claim 1, wherein the quotient of the pressure ratio over the permeate separation stage to the pressure ratio over the feed stream separation stage is in a range of from 1 to 3.

Ex. 1001, Priske-'380, claim 10

Claim 10 – The method of claim 1, wherein the quotient of the pressure ratio over the permeate separation stage to the pressure ratio over the feed stream separation stage is in a range of from 1 to 3.

See [1.f.iii], *supra*. Scholz's Figure 7 discloses a range for the 3/1 pressure ratio quotient of 0.42-0.77, which does not overlap with the 1-3 claimed range, but the high-end of the Scholz range is close to the low-end of the claimed range. AP-1003, ¶¶212-213, 236-237.

Corrected Petition, 57

Claim 10 | Scholz Does Not Disclose a “Range”

212. In Scholz’s Figure 7 (a) system, “the quotient of the pressure ratio over the permeate separation stage to the pressure ratio over the feed stream separation stage” (“3/1 pressure ratio quotient”) is $0.77 = ((3.5 \text{ bar} \div 1 \text{ bar}) \div (16 \text{ bar} \div 3.5 \text{ bar}))$. A POSITA would have understood system (a) in Scholz’s Figure to meet limitation [1f.iii] of claim 1 of the ‘380 patent.

213. The 3/1 pressure ratio quotient in system (b) in Scholz’s Figure 7 is $0.42 = ((2.6 \text{ bar} \div 1 \text{ bar}) \div (16 \text{ bar} \div 2.6 \text{ bar}))$. The range between the values in Scholz’s systems (a) and (b) (0.42-0.77) overlaps the 0.5-8 range in limitation [1f.iii] of the ‘380 patent.

Ex. 1003, Baker Decl., ¶¶ 212-213

Claim 10 | Scholz Does Not Render Obvious the Claimed Range



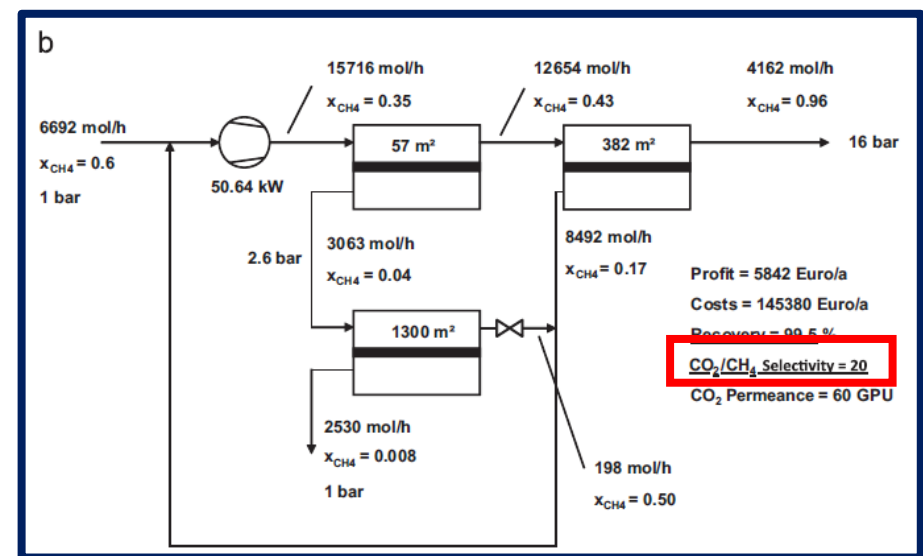
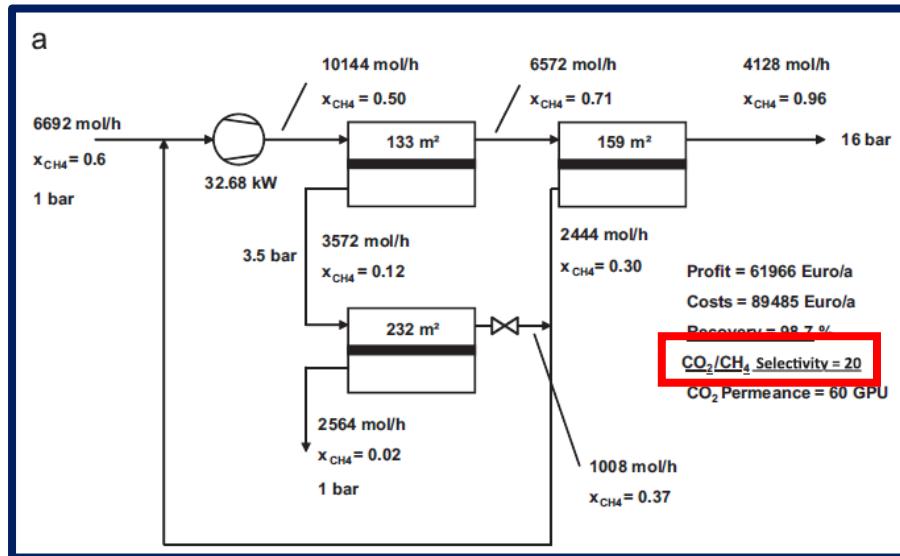
134. Further, Dr. Baker argues again that a person of ordinary skill would arrive at the claimed values of 1 to 3 by mere experimentation, starting from the values disclosed in Scholz. I disagree. Modifying the closest value of Scholz to arrive at the closest end of the claimed range would constitute a 30% increase in the pressure ratio quotient ($[1-0.77]/0.77$). Again, while a person of ordinary skill can manipulate processes using programs such as Aspen, ChemCad, or GAMS, changing pressure ratios by 30% is more than mere experimentation, it is more like reworking the entire system. Such a change to the pressure ratio would require significant changes to other process parameters too. Since the process in Scholz 7(a) was shown to achieve its required purity of at least 95% and optimized for the highest profit, a person of ordinary skill would understand this process works for its intended purpose and have no motivation to modify it.

Ex. 2008, Geise Decl., ¶ 134

Claim 12 | Scholz Does Not Render Obvious the Claimed Range

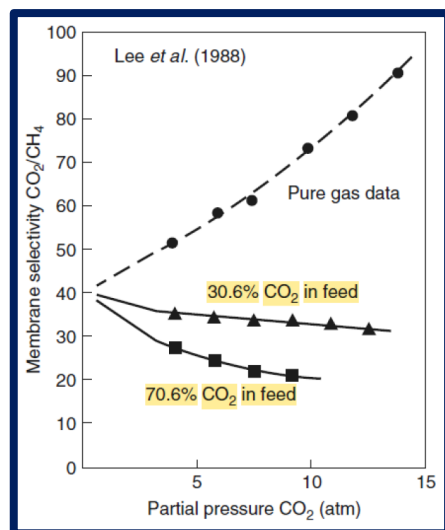
12. The method of claim 11, wherein the gas separation membranes in the feed stream separation stage have a pure gas selectivity for CO₂/CH₄ of from 20 to 100.

Ex. 1001, Priske-'380, claim 12



Ex. 1007, Scholz, Figures 7(a), 7(b)

Dependent Claim 12 is Not Obvious



Ex. 1013, Baker Textbook, Figure 8.8 (annotated)

- No data for 50% CO_2 stream as in Scholz Figure 7(a), or 65% CO_2 stream as in Scholz Figure 7(b).

Ex. 1013, Baker Textbook, 345, Figure 8.8

Ex. 1007, Scholz, Figures 7(a), 7(b)

OSI Pharms., LLC v. Apotex Inc.

Reversing Board's determination of obviousness where references lacked "any data or other information" regarding the proposed combination because "[m]ere speculation" is not substantial evidence."

939 F.3d 1375, 1382, 1385 (Fed. Cir. 2019)

Scholz Does Not Teach Claim 17



17. The method of claim 2, wherein the compressor is a multistage compressor and the second permeate stream, the third retentate stream, or both are introduced into the compressor between two compression stages.

Q. And generally speaking, is a multistage compressor more expensive than a one stage compressor?

MR. LANE: Objection. Form.

A. Multistage implies that the compressor is built to go from — from low pressure to a fairly high pressure, so it will be a more expensive machine than a single stage. Trying to do a big pressure ratio in a single stage, people do it, but not — not for any reasonable sized machine.

Ex. 2009, Baker Dec. Dep., 109:14-22

Scholz Does Not Teach Claim 17



17. The method of claim 2, wherein the compressor is a multistage compressor and the second permeate stream, the third retentate stream, or both are introduced into the compressor between two compression stages.

139. A person of ordinary skill would understand that compressors are a significant cost component of a membrane gas separation system and would not simply seek to introduce a multistage compressor without explicit motivation. Further, since the recycle streams shown in Scholz are combined with the crude gas stream that is at or near atmospheric pressure, it is assumed the recycle streams are also at or near atmospheric pressure. **Thus, there would be no need for a multistage compressor since the crude and recycle streams are at or are nearly at the same pressure.** Finally, if the pressure of the recycle streams (or the crude stream) was raised to enable use of a multistage compressor, this would directly impact the pressure ratios of the system (and the quotients of them), thus significantly modifying the process.

Ex. 2008, Geise Decl., ¶ 139

Petitioner Has Not Met Its Burden for Grounds 1 or 2

Ground 1 Contains Several Fatal Flaws

In Ground 1, Petitioner is requesting the Board:

- 1. Look past statutory requirements and fix Petitioner's errors in its alleged prior art bases**
- 2. Accommodate new theories relying on "intuition" that Petitioner's expert admitted are incorrect**
- 3. Ignore Ungerank's express teachings regarding the capacities of the membranes used therein**

Ground 2 Similarly Fails

In Ground 2, Petitioner has:

- 1. Stopped arguing this ground entirely in its Reply**
- 2. Ignored claim construction entirely, in the face of a defined term in the Priske-'380 Patent**
- 3. Tortuously interpreted Scholz to teach “ranges” by looking at values in two distinct processes**

Index

• Overview of the Technology.....	2
• The Challenged Patent.....	5
• The Asserted Prior Art.....	23
• Overview of the Grounds.....	36
• The Disputed Issues.....	38
– Ground 1.....	40
• Ungerank Is Not Prior Art.....	41
• Petitioner’s Original Obviousness Theory Is Incorrect.....	49
• Petitioner’s New Obviousness Theory Is Also Incorrect.....	68
• Petitioner Mischaracterizes the Problem to Be Solved.....	75
• No Rationale to Start With Ungerank CE1.....	83
• No Reasonable Expectation of Success.....	90
• Claims 9 and 12 are not Obvious.....	101
– Ground II.....	106
• Conclusion.....	128